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(54) MATERIAL FOR LIGHT-EMITTING DEVICE AND LIGHT-EMITTING DEVICE USING IT

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a light-emitting device excellent in heat stability, high in utilization efficiency of electric energy and color purity.

SOLUTION: The material for the light-emitting device comprises a compound having an oligopyridine skeleton formed by connecting three or more pyridine rings through single bonds, the compound being structured only by covalent bonds.

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CLAIMS

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[Claim(s)]

[Claim 1]A charge of light emitting device material containing a compound which has the oligo pyridine skeleton with which three or more pyridine rings were connected in a single bond, and does not include combination of those other than a covalent bond.

[Claim 2]The charge of light emitting device material according to claim 1, wherein said compound is a thing containing two or more oligo pyridine skeletons.

[Claim 3]The charge of light emitting device material according to claim 2, wherein two or more oligo pyridine skeletons are connected by at least one connecting group of a conjugated bond, a multivalent aromatic hydrocarbon group, and a multivalent aroma heterocycle group.

[Claim 4]The charge of light emitting device material according to claim 1, wherein an oligo pyridine skeleton is the Tarr pyridine skeleton.

[Claim 5]A light emitting device using the charge of light emitting device material according to claim 1 into this element in a light emitting device which a luminous layer having contained photogene is made to intervene between the anode and the negative pole, and emits light with electrical energy.

[Claim 6]The light emitting device according to claim 5 by which making an electron transport layer intervene between a luminous layer and the negative pole, and said charge of light emitting device material being included in this electron transport layer.

[Claim 7]The light emitting device according to claim 5 constituting a display which a light emitting device displays with a matrix and/or a segment system.

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[Translation done.]

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention]This invention is an element which can change electrical energy into light, and relates to the charge of light emitting device material and a light emitting device available in the field of a display device, a flat-panel display, a back light, lighting, interior design, a sign, a signboard, an electro photography machine, a lightwave signal generator, etc.

[0002]

[Description of the Prior Art]Research of the organic laminated thin film light emitting device of emitting light when the electron poured in from the negative pole and the electron hole poured in from the anode recombine within the organic fluorescent substance inserted into two poles is done actively in recent years. The bright luminescence under a thin shape and low driver voltage and multicolor luminescence by choosing a fluorescent material are the features, and this element attracts attention.

[0003]Since this research showed that an organicity [ C.W.Tang and others of Kodak ] laminated thin film element emitted light to high-intensity, the research institution of (Appl. Phys. Lett., 51 (12) 21, p.913 (1987)) and many is inquiring. The typical composition of the organic laminated thin film light emitting device which the research consortium of Kodak presented, Mg:Ag was provided one by one on the ITO glass substrate as the diamine compound of electron hole transportability, 8-hydroxy kino RINARU minium which is luminous layers, and the negative pole, and the green emission of  $1000\text{cd}/\text{m}^2$  was possible at about [ 10V ] driver voltage.

[0004]Since it is possible to obtain the various luminescent color by using various fluorescent materials for a luminous layer, the utilization research to a display etc. is prosperous. In the trichromatic luminescent material, research of green emission material is progressing most and research is wholeheartedly made in red light material and blue light material now aiming at the improvement in the characteristic.

[0005]About the composition of this organic laminated thin film light emitting device, what provided suitably the electron transport layer other than the above-mentioned anode / electron hole transporting bed / luminous layer / negative pole is known. With an electron hole transporting bed, it has the function to convey the electron hole poured in from the anode to a luminous layer, and one electron transport layer conveys the electron poured in from the negative pole to a luminous layer. It is known by inserting these layers between a luminous layer and two poles that luminous efficiency and endurance will improve. As an example of the element composition using these, the anode / electron hole transporting bed / luminous layer / electron transport layer / negative pole, the anode / luminous layer / electron transport layer / negative pole, etc. are mentioned, and research of the organic compound suitable for each class is done centering on the hole transporting material.

[0006]

[Problem(s) to be Solved by the Invention]However, conventionally, the desired luminescent color was not obtained for the reason of causing a luminescent material and an interaction, even if an electron transport material is seldom studied but it uses a few existing materials, or luminescence of the electron transport material itself being intermingled, and there were

problems, like the endurance of that from which efficient light is obtained is low. For example, although a specific phenanthroline derivative shows efficient light, it crystallizes by prolonged energization and it has a problem which a thin film nebula-izes. Although there are a quinolinol metal complex and a benzoquinolinol metal complex to show the characteristic comparatively good for luminous efficiency and endurance, Since these have high bluish green – the luminescence capability in yellow in this material itself, when it uses as an electron transport material, there is a possibility that luminescence of material itself [ these ] may be intermingled and color purity may get worse. Although there is an example which used the diquinolin-derivative and the Tori quinoline derivative for JP,7-150137,A, It is unknown whether although endurance shows the comparatively good characteristic, it is unstated about improvement in luminous efficiency, and it functions as an electron transport material in luminescence by the side of yellow – long wavelength like red light.

[0007]Although there is an example which thin-film-ized the metal complex of a bipyridine or TAPI lysine by the wet method to JP,2001-226146,A, and was used for the light emitting device, application is thermally impossible for an unstable and dry process, and also luminescent ability is also low.

[0008]This invention solves the problem of this conventional technology, and is excellent in thermal stability, its luminous efficiency is high and an object of this invention is to provide the light emitting device using the charge of light emitting device material and it which make possible the light emitting device excellent in color purity.

[0009]

[Means for Solving the Problem]This invention is a charge of light emitting device material being a compound which has the oligo pyridine skeleton with which three or more pyridine rings were connected in a single bond, and does not include combination of those other than a covalent bond.

[0010]It is a light emitting device furthermore characterized by this invention using the above-mentioned charge of light emitting device material into this element in a light emitting device which a luminous layer having contained photogene is made to intervene between the anode and the negative pole, and emits light with electrical energy.

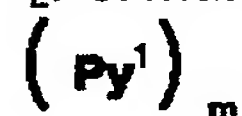
[0011]

[Embodiment of the Invention]The compound which has the oligo pyridine skeleton which connected the pyridine ring of this invention in or more three single bond is explained in detail. The oligo pyridine skeleton which connected the pyridine ring in or more three single bond is expressed with a following general formula (1).

General formula (1)

[0012]

[Formula 1]

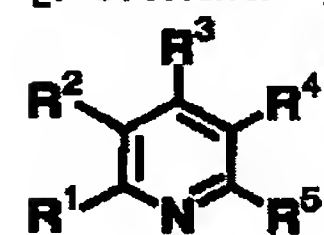


[0013]Here,  $\text{Py}^1$  expresses the pyridine ring expressed with a following general formula (2). m is three or more integers.

General formula (2)

[0014]

[Formula 2]



[0015]here,  $\text{R}^1 - \text{R}^5$  may be the same, or may differ from each other, at least two are [ pyridyl group / of an end ] a single bond about the pyridyl group in one and a chain, and the remainder, Hydrogen, an alkyl group, a cycloalkyl group, an aralkyl group, an alkenyl group, A cycloalkenyl group, an alkynyl group, a hydroxyl group, a sulfhydryl group, an alkoxy group, An alkylthio group,



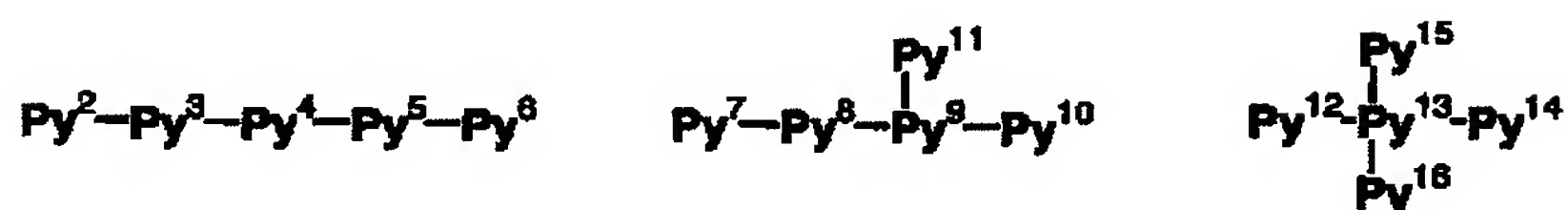
an aryl ether group, an arylthio ether group, an aryl group, A heterocycle group, halogen, haloalkane, a halo alkene, a halo alkyne, As being chosen from the condensed rings formed between a cyano group, an aldehyde group, a carbonyl group, a carboxyl group, an ester group, a carbamoyl group, an amino group, a nitro group, a silyl group, a siloxanyl group, and a contiguity substituent, it is a kind as it is few.

[0016]An alkyl group shows saturated-aliphatic-hydrocarbon groups, such as a methyl group, an ethyl group, a propyl group, and a butyl group, among these substituents, and even if this is replaced also in no replacing, it is not cared about. A cycloalkyl group shows saturated alicyclic hydrocarbon groups, such as for example, cyclopropyl, cyclohexyl, norbornyl, and adamantyl, and even if this is replaced also in no replacing, it is not cared about. An aralkyl group shows an aromatic hydrocarbon group through aliphatic hydrocarbon, such as benzyl and a phenylethyl group, and even if each of aliphatic hydrocarbon and aromatic hydrocarbon is replaced also in no replacing, it is not cared about. An alkenyl group shows an unsaturation aliphatic hydrocarbon group including double bonds, such as a vinyl group, an allyl group, and a butadienyl group, and even if this is replaced also in no replacing, it is not cared about. A cycloalkenyl group shows an unsaturation alicyclic hydrocarbon group including double bonds, such as for example, a cyclopentenyl group, a cyclopentadienyl group, and a cyclohexene group, and even if this is replaced also in no replacing, it is not cared about. An alkynyl group shows an unsaturation aliphatic hydrocarbon group including triple bonds, such as for example, an acetylenyl group, and even if this is replaced also in no replacing, it is not cared about. An alkoxy group shows an aliphatic hydrocarbon group through ether bonds, such as a methoxy group, and even if an aliphatic hydrocarbon group is replaced also in no replacing, it is not cared about. An oxygen atom of an ether bond of an alkoxy group is replaced by an alkylthio group by sulfur atom. An aryl ether group shows an aromatic hydrocarbon group through ether bonds, such as a phenoxy group, and even if an aromatic hydrocarbon group is replaced also in no replacing, it is not cared about. An oxygen atom of an ether bond of an aryl ether group is replaced by an arylthio ether group by sulfur atom. An aryl group shows aromatic hydrocarbon groups, such as a phenyl group, a naphthyl group, a biphenyl group, a phenanthryl group, a terphenyl group, and a pyrenyl group, and even if this is replaced also in no replacing, it is not cared about. A heterocycle group shows a cyclic structure group which has atoms other than carbon, such as for example, a furanyl group, a thiophenyl group, an oxazolyl group, a pyridyl group, a quinolinyl group, and a carbazolyl group, and even if this is replaced also in no replacing, it is not cared about. Halogen shows fluoride, chlorine, bromine, and iodine. A part or all of the above-mentioned alkyl groups, such as a trifluoromethyl group, an alkenyl group, and an alkynyl group shows what was replaced with the above-mentioned halogen, and even if haloalkane, a halo alkene, and a halo alkyne are replaced also in no replacing, they do not care about the remaining portion. To an aldehyde group, a carbonyl group, an ester group, a carbamoyl group, and an amino group, aliphatic hydrocarbon, Including what was replaced by alicyclic hydrocarbon, aromatic hydrocarbon, heterocycle, etc., further, even if aliphatic hydrocarbon, alicyclic hydrocarbon, aromatic hydrocarbon, and heterocycle are replaced also in no replacing, they are not cared about. A silyl group shows silicon compound groups, such as for example, a trimethylsilyl group, and even if this is replaced also in no replacing, it is not cared about. A siloxanyl group shows a silicon compound group through ether bonds, such as for example, a trimethyl siloxanyl group, and even if this is replaced also in no replacing, it is not cared about. If said general formula (2) explains a condensed ring formed between contiguity substituents, it will form a condensed ring of a pyridyl ring, conjugate, or non-conjugate by a part of  $R^1$ ,  $R^2$  and  $R^2$ ,  $R^3$  and  $R^3$ ,  $R^4$ , or  $R^4$  and  $R^5$ . And these condensed rings may contain nitrogen, oxygen, and a sulfur atom in endocyclic structure, and may condense them with another ring.

[0017]In a general formula (1), although m pyridine rings are connected in a single bond, they may lead to straight chain shape, or may branch. For example, in the case of m= 5, the following three kinds of structures are mentioned.

[0018]

[Formula 3]



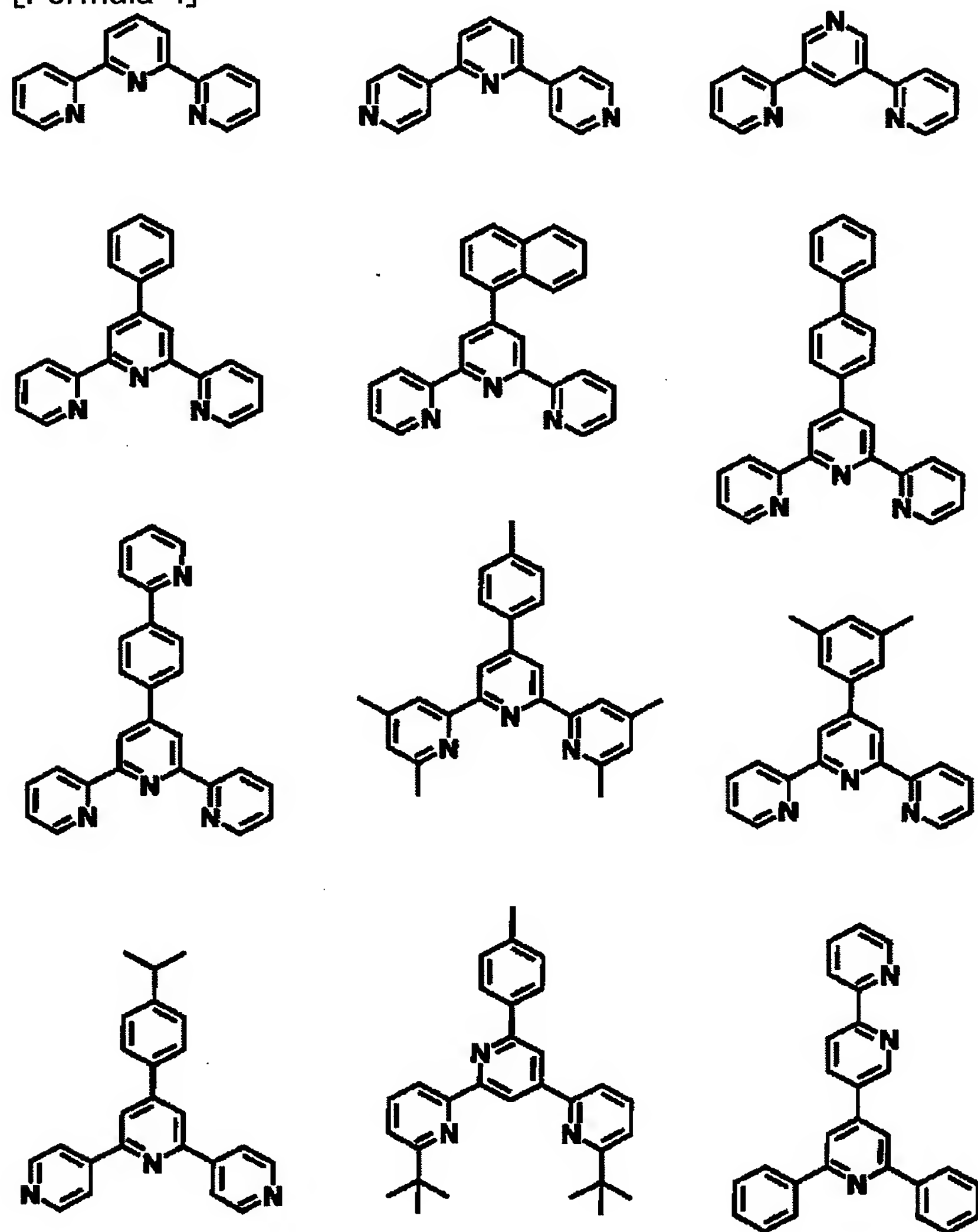
[0019] Here,  $\text{Py}^2 - \text{Py}^{16}$  are pyridine rings expressed with the same general formula (2) that may differ but.

[0020] The compound expressed with a general formula (1) comprises only a covalent bond. When a coordinate bond etc. are used, for example except a covalent bond, the thermal stability of a compound is low and is not preferred as a charge of light emitting device material.

[0021] Although you may connect by any part of a pyridine ring as a connection part of the pyridine rings of a general formula (1), when a composite ease etc. are taken into consideration, it is preferred to have connected by the 2nd place of a pyridine ring, the 4th place, and the 6th place. The following is mentioned as an example of a general formula (1).

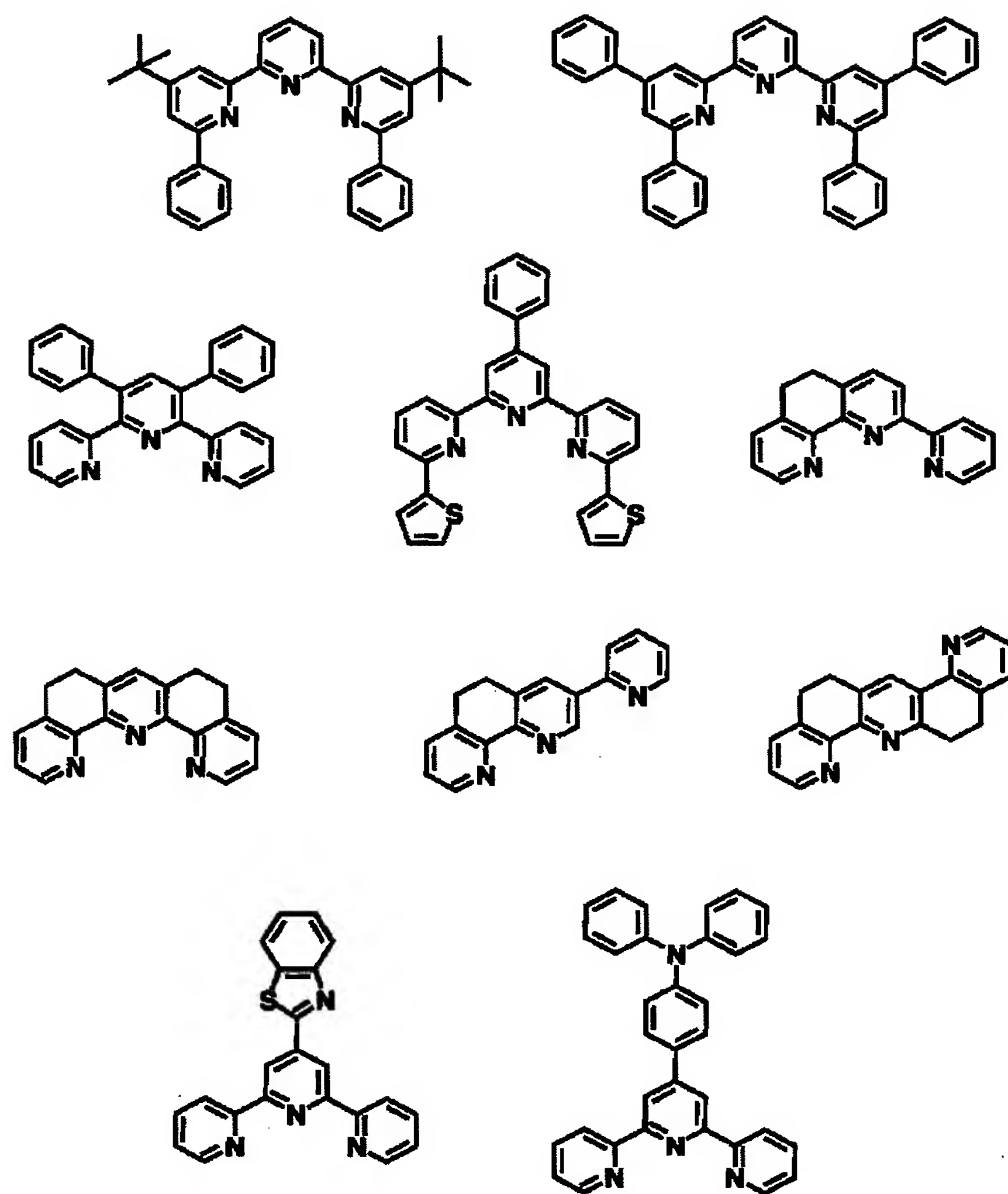
[0022]

[Formula 4]

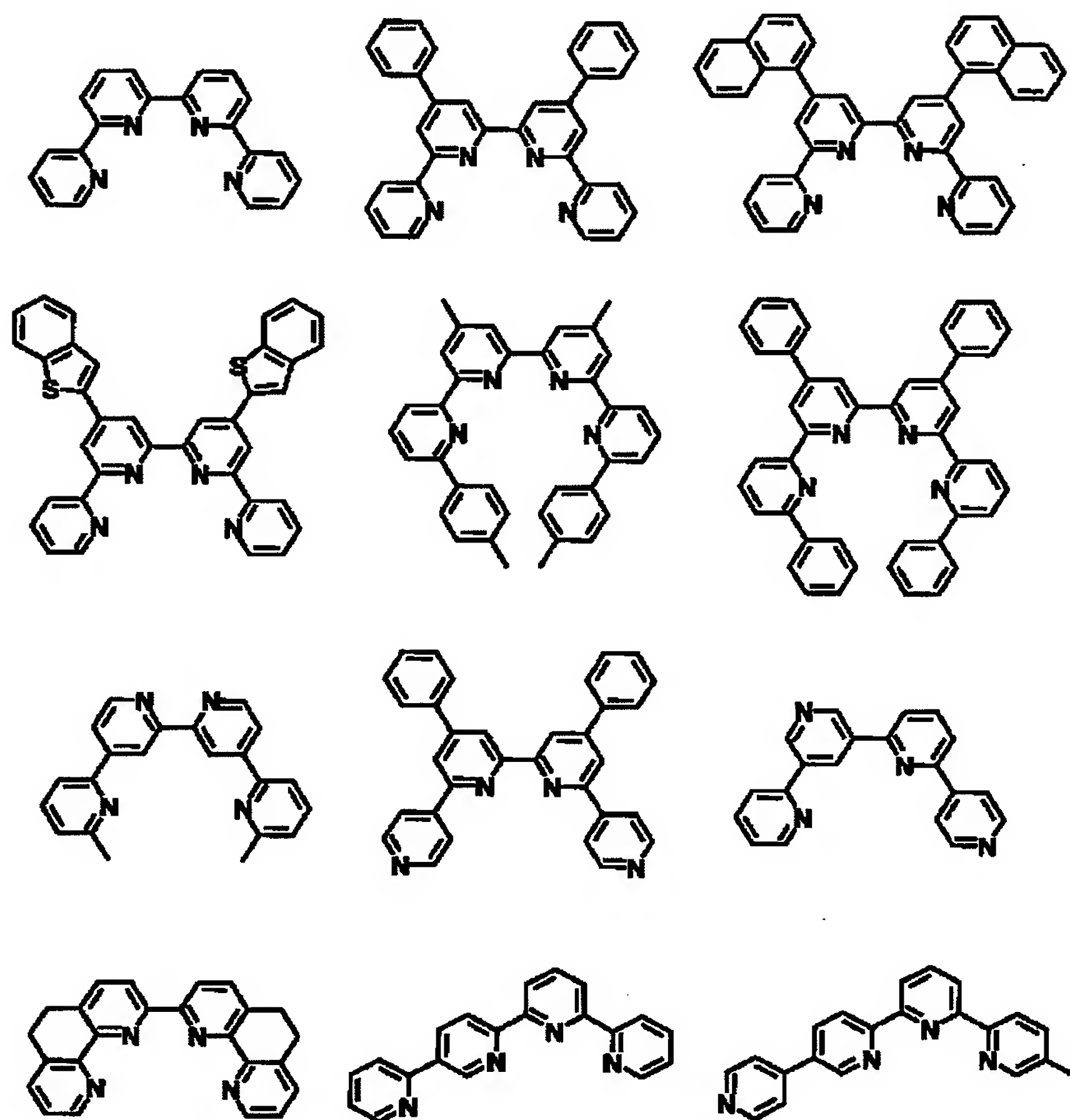


[0023]

[Formula 5]



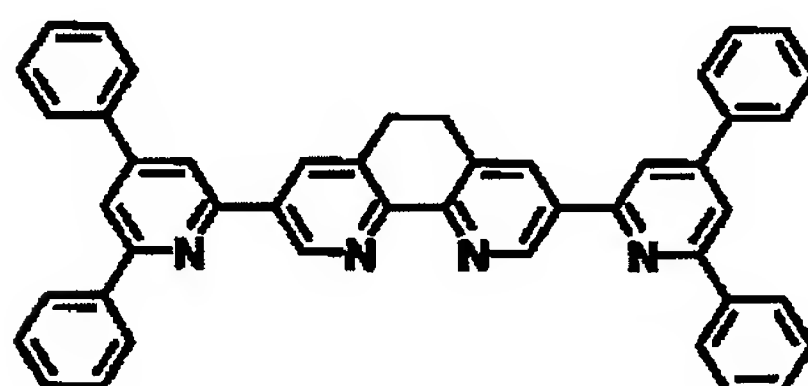
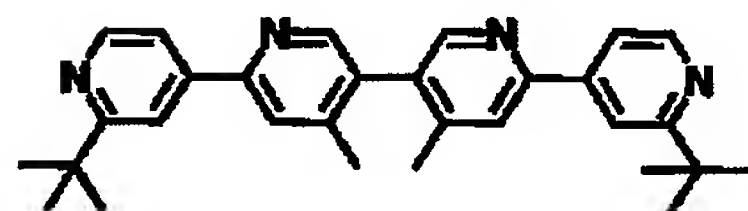
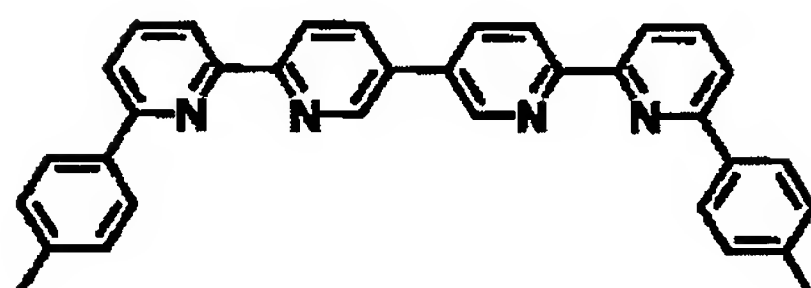
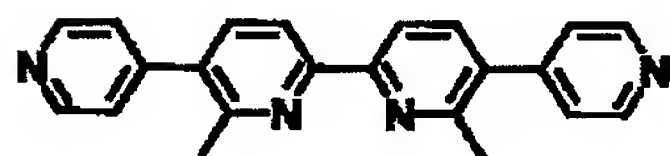
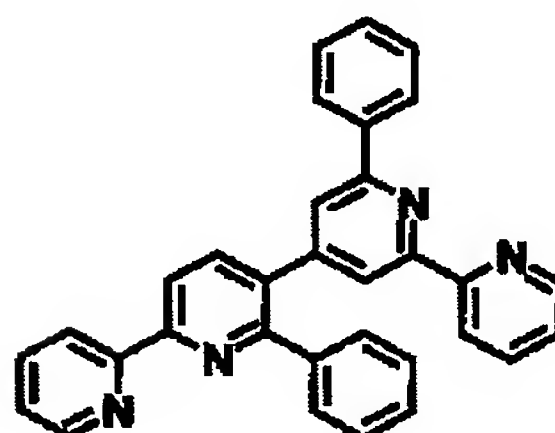
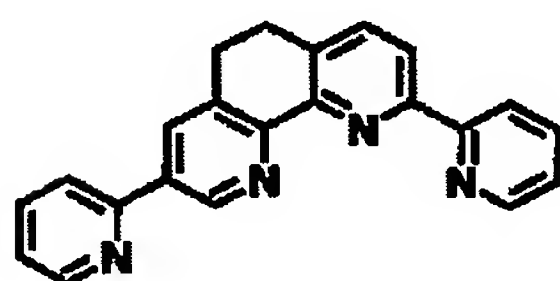
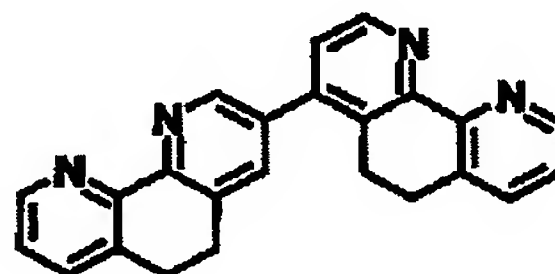
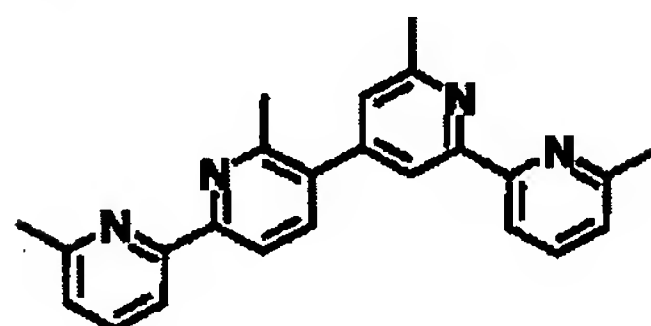
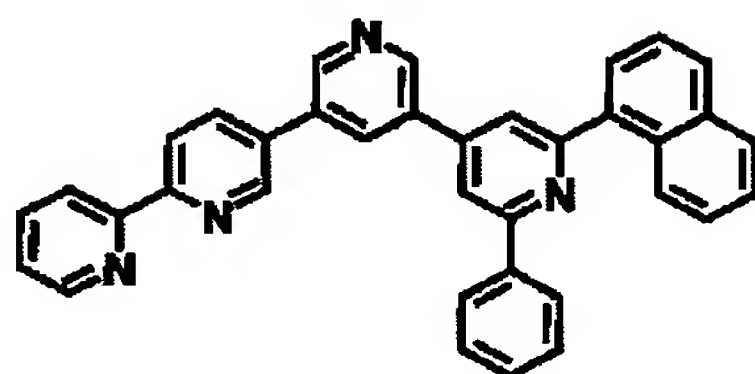
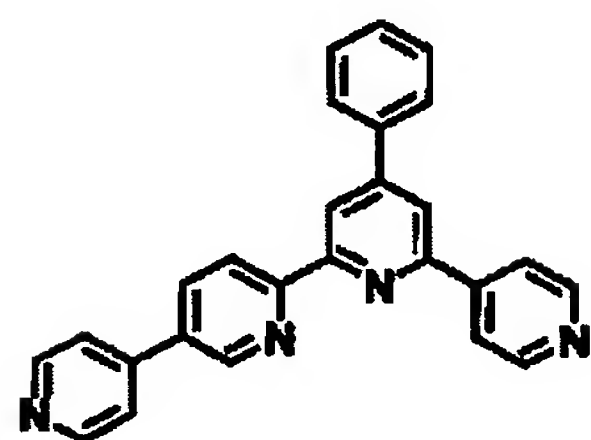
[0024]  
[Formula 6]



[0025]

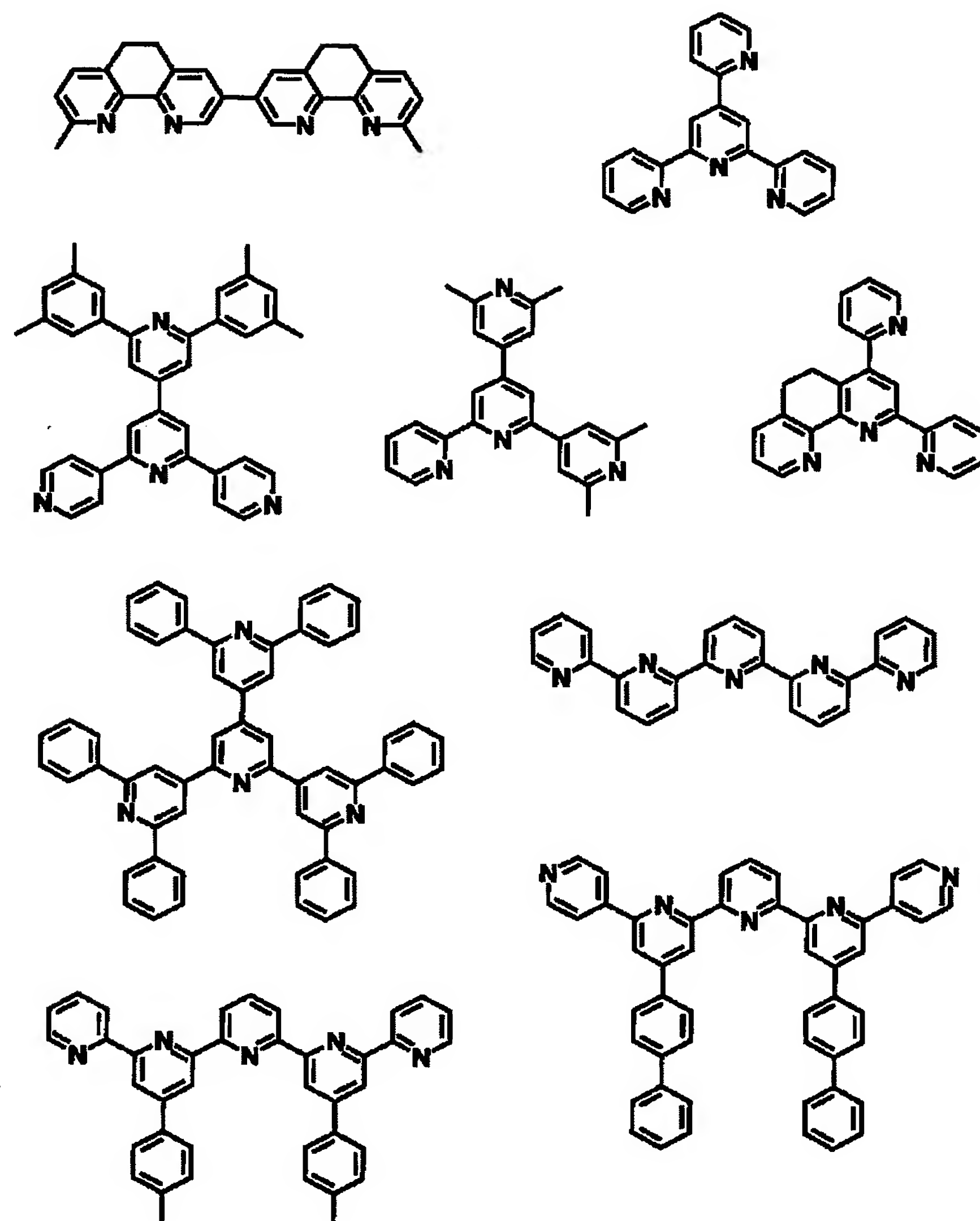
[Formula 7]





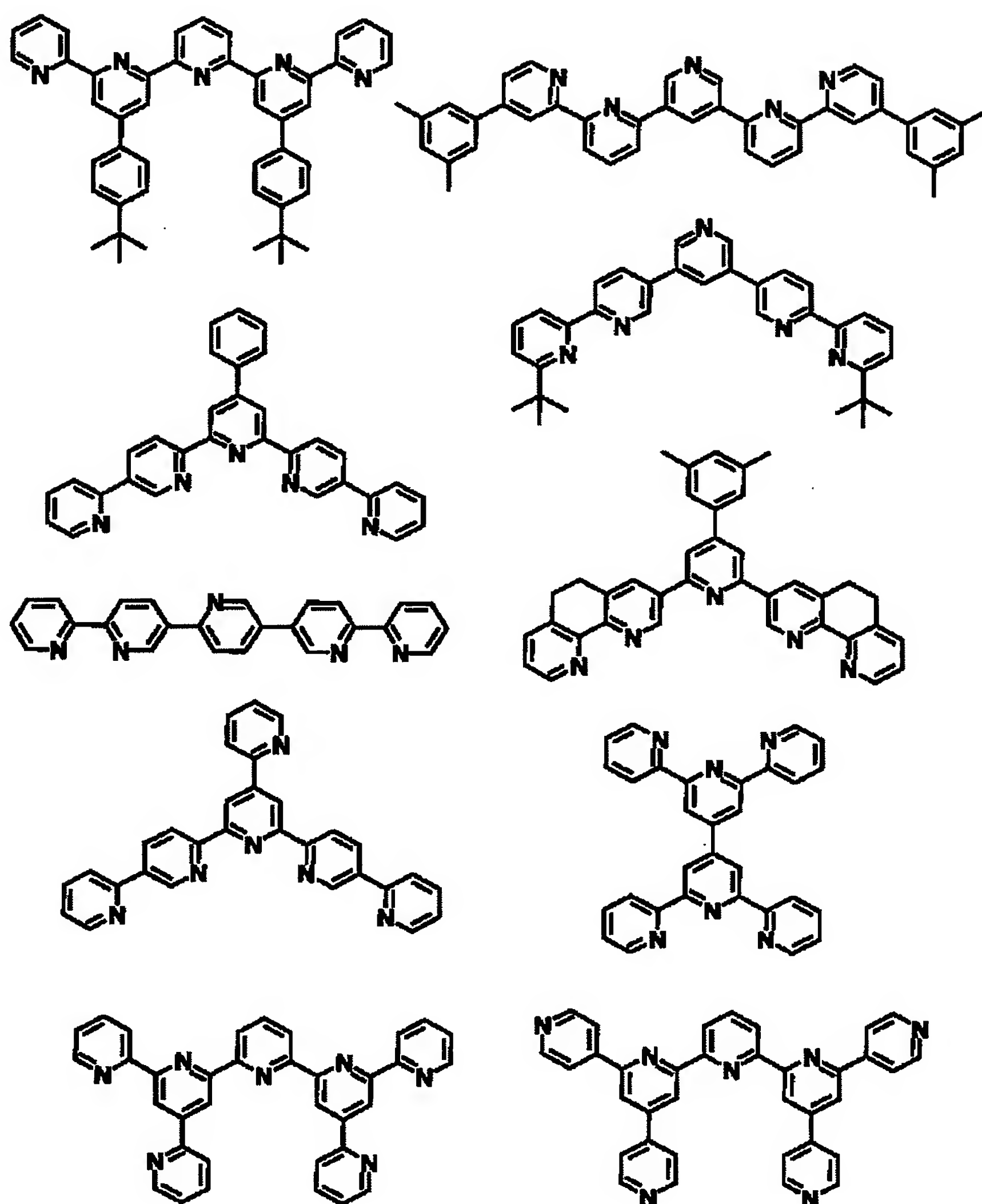
[0026]

[Formula 8]



[0027]

[Formula 9]



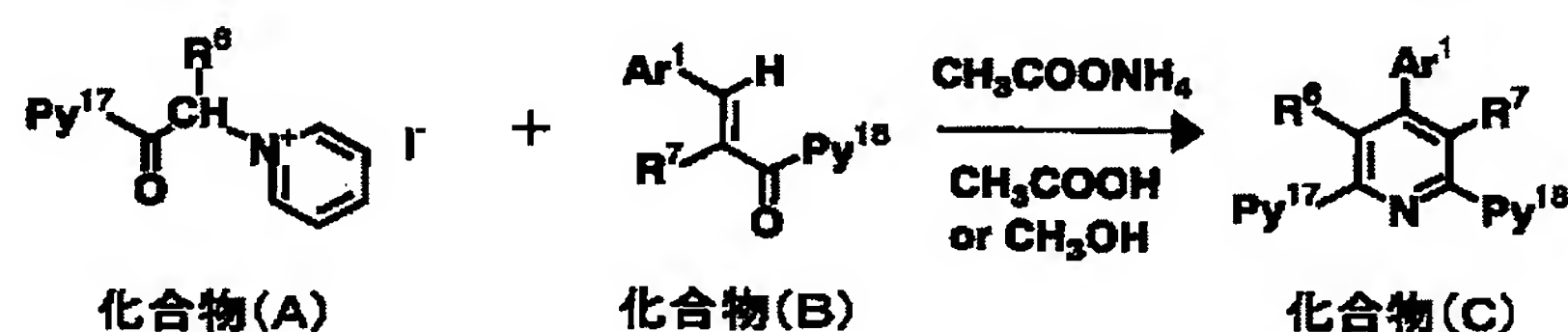
[0028] Although the compound expressed with a general formula (1) is compoundable by the following methods, for example, it is not limited to this.

[0029] composition of oligo pyridine -- as a reference -- Synthesis, the 1st (1976) page and Coordination Chemistry Reviews, and vol.160 (1997) -- the 1st page etc. are mentioned. For example, a TAPI lysine (oligo pyridine which pyridine ring connected in 3 single bonds) skeleton expressed with a compound (C), A compound (A) obtained from acetylpyridine, and iodine and pyridine and a compound (B) obtained from a condensation reaction of acetylpyridine and aryl aldehyde are made to react in bottom acetic acid of ammonium acetate existence, or methanol, and it can compound by processing with a conventional method (scheme I).

[0030]

[Formula 10]

(スキーム I)



[0031] Here, explanation of R<sup>6</sup> and R<sup>7</sup> is the same as that of the substituent explained in the above-mentioned R<sup>1</sup> - R<sup>5</sup>. Ar<sup>1</sup> expresses the aryl group which is not replaced [ substitution or ]. Py<sup>17</sup> and Py<sup>18</sup> are pyridine rings which may be the same, or may differ and are expressed with a

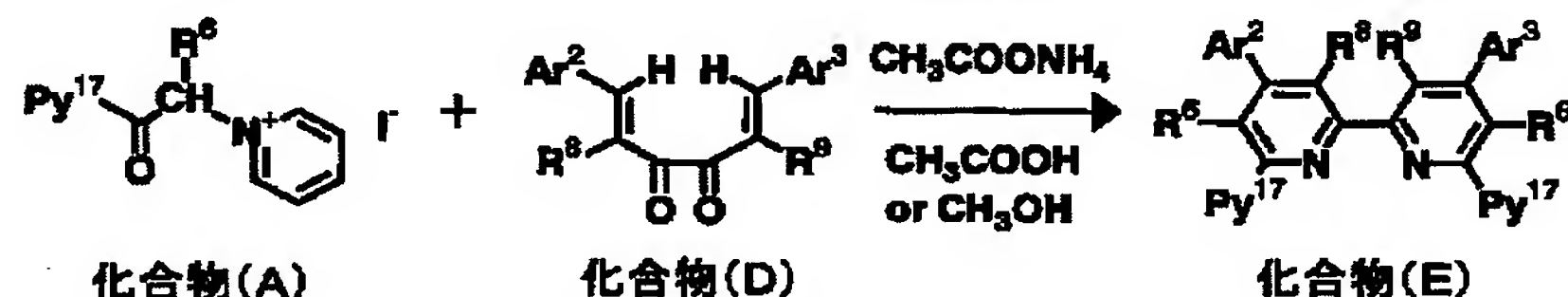
general formula (2).

[0032] An oligo pyridine skeleton whose number of pyridine rings is four or more is compoundable using the same synthetic method as the above-mentioned Tarr pyridine skeleton. A compound (E) which a pyridine ring connected in 4 single bonds when a compound (D) was used instead of a compound (B), and (quarter pyridine) For example, the (scheme II), If a compound (F) is used instead of a compound (A), a pyridine ring can compound a compound (G) connected in 5 single bonds, and (thornhead pyridine) (scheme III). If a compound (H) is used instead of a compound (F) and a compound (B) instead of a compound (A), a pyridine ring can compound compound (I) (SEPUTI pyridine) connected in 7 single bonds (scheme IV).

[0033]

[Formula 11]

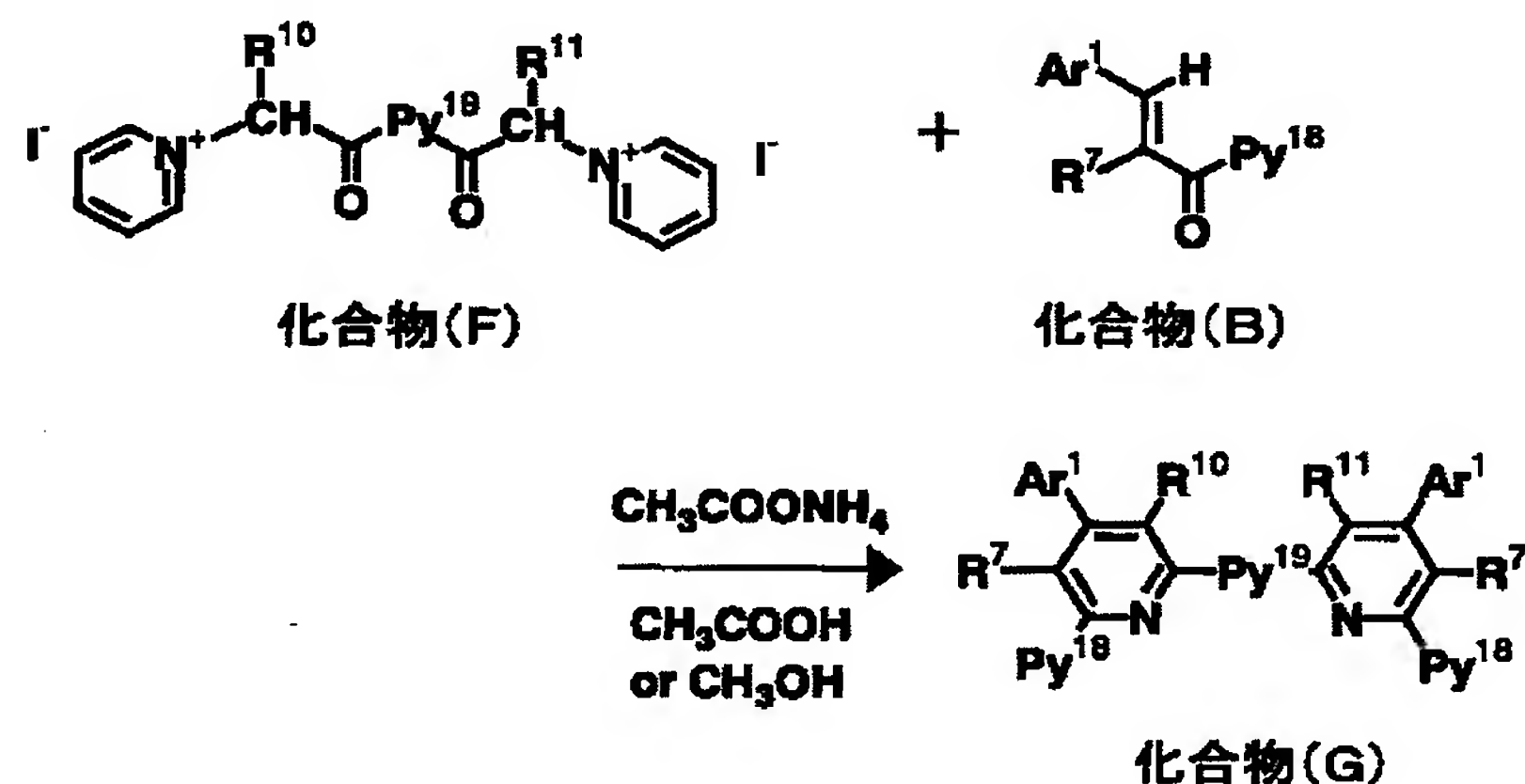
(スキーム II)



[0034]

[Formula 12]

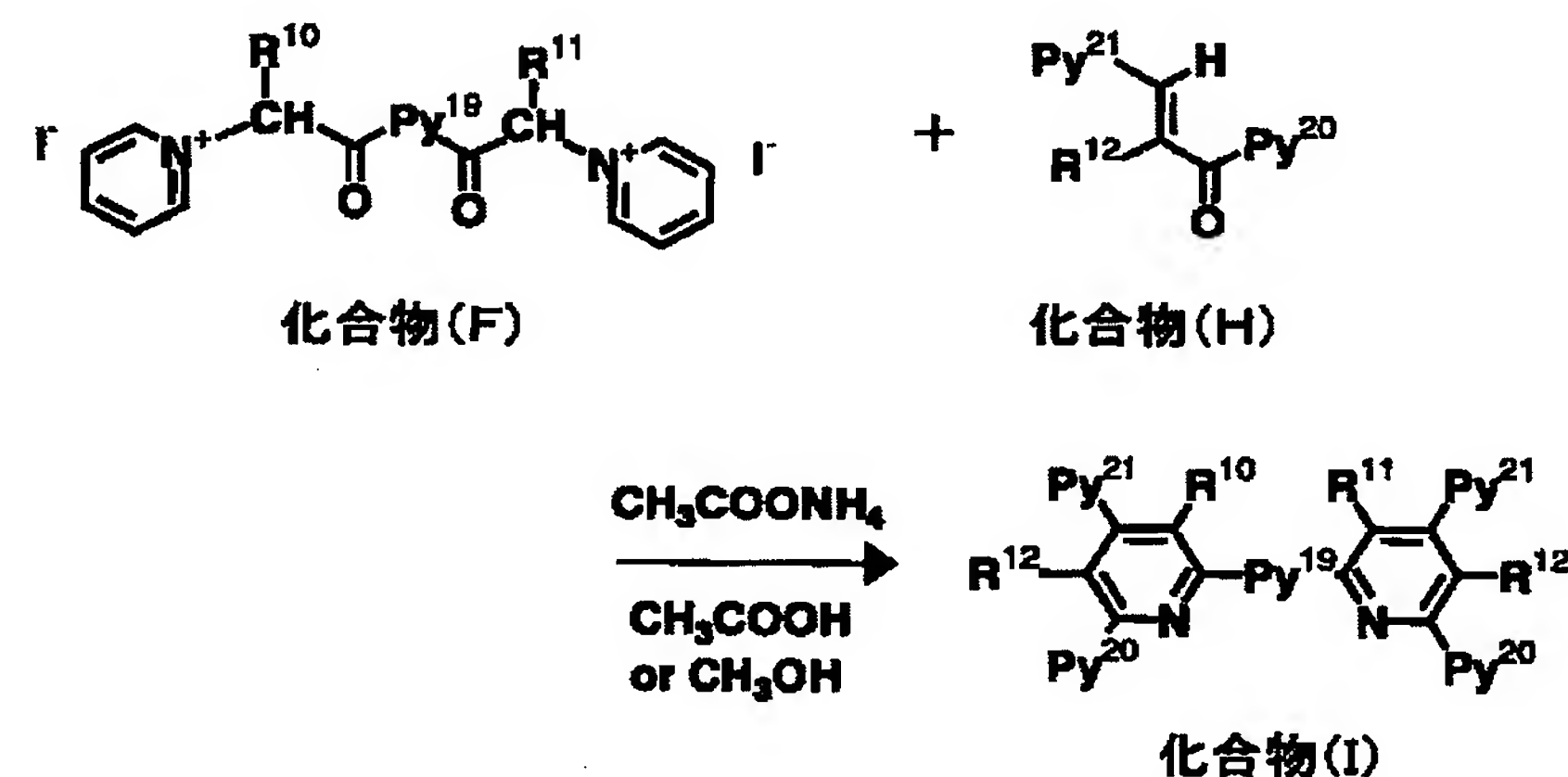
(スキーム III)



[0035]

[Formula 13]

(スキーム IV)



[0036] Here, explanation of  $R^8 - R^{12}$  is the same as that of the substituent explained in the above-mentioned  $R^1 - R^5$ .  $Ar^2$  and  $Ar^3$  may be the same, or may differ from each other, and

express the aryl group which is not replaced [ substitution or ].  $\text{Py}^{19} - \text{Py}^{21}$  are pyridine rings which may be the same, or may differ and are expressed with a general formula (2).

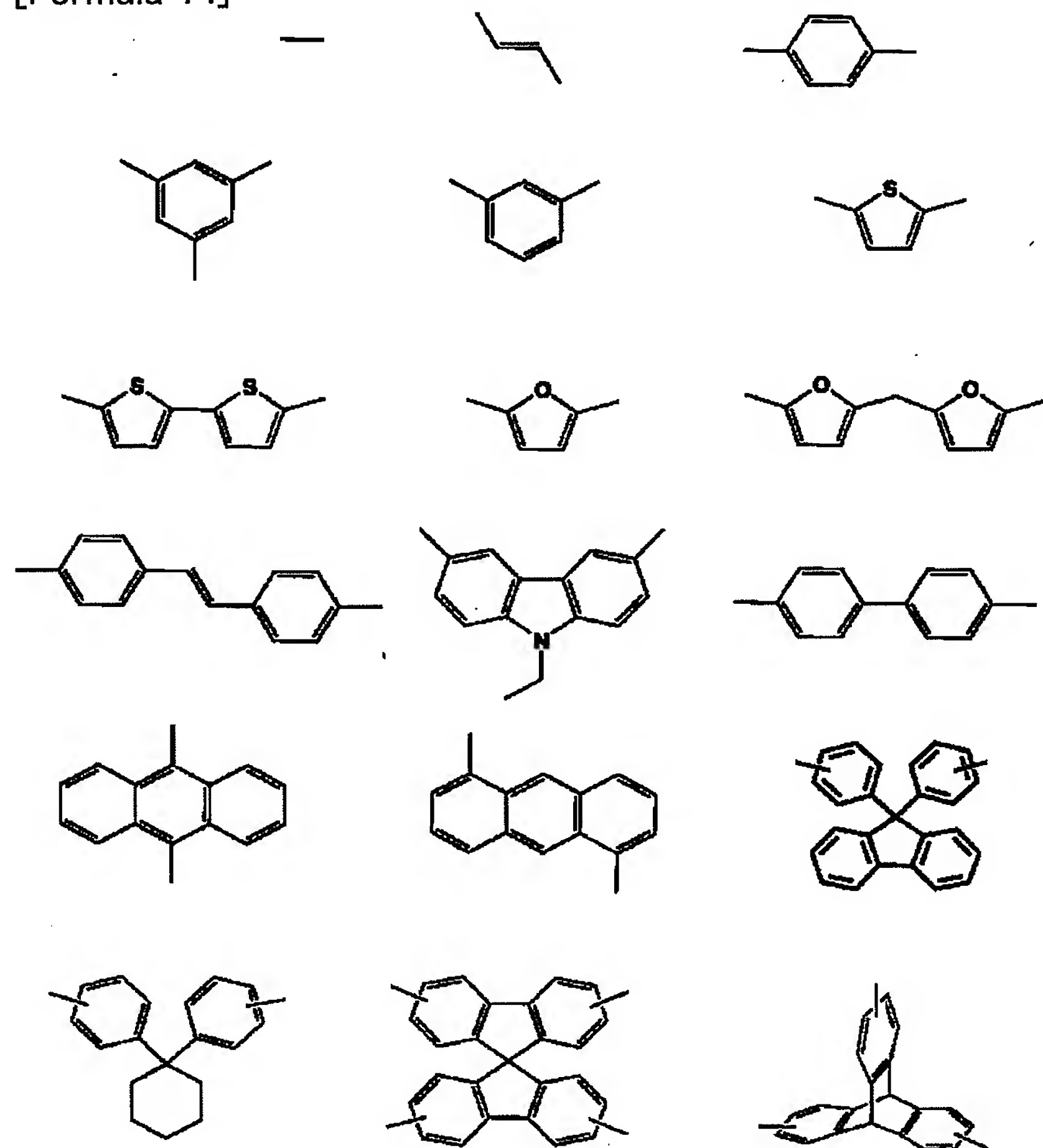
[0037] Considering the ease of receiving of material, and the eases of composition and refining, the compound which has the Tarr pyridine skeleton also in the above-mentioned oligo pyridine skeleton is preferred.

[0038] In order to obtain stable luminescence over a long time, a material excellent in thermal stability or thin-film-forming nature is desired, and a compound which has two or more oligo pyridine skeletons also in a compound which has an oligo pyridine skeleton can be raised as a more desirable example. About explanation of an oligo pyridine skeleton, it is the same as that of what was mentioned above.

[0039] In order to obtain bright luminescence using a compound in this invention, it is preferred that electron transportation ability uses a high compound. Then, it is preferred to use a compound which has two or more oligo pyridine skeleton structures where said three or more pyridine rings were connected in a single bond and in which two or more oligo pyridine skeleton structures are connected by at least one of a conjugated bond, a multivalent aromatic hydrocarbon group, and the multivalent aroma heterocycle groups. Even if a starting connecting group has a substituent and it does not have it, it is not cared about. The following structures are mentioned as an example of a starting connecting group.

[0040]

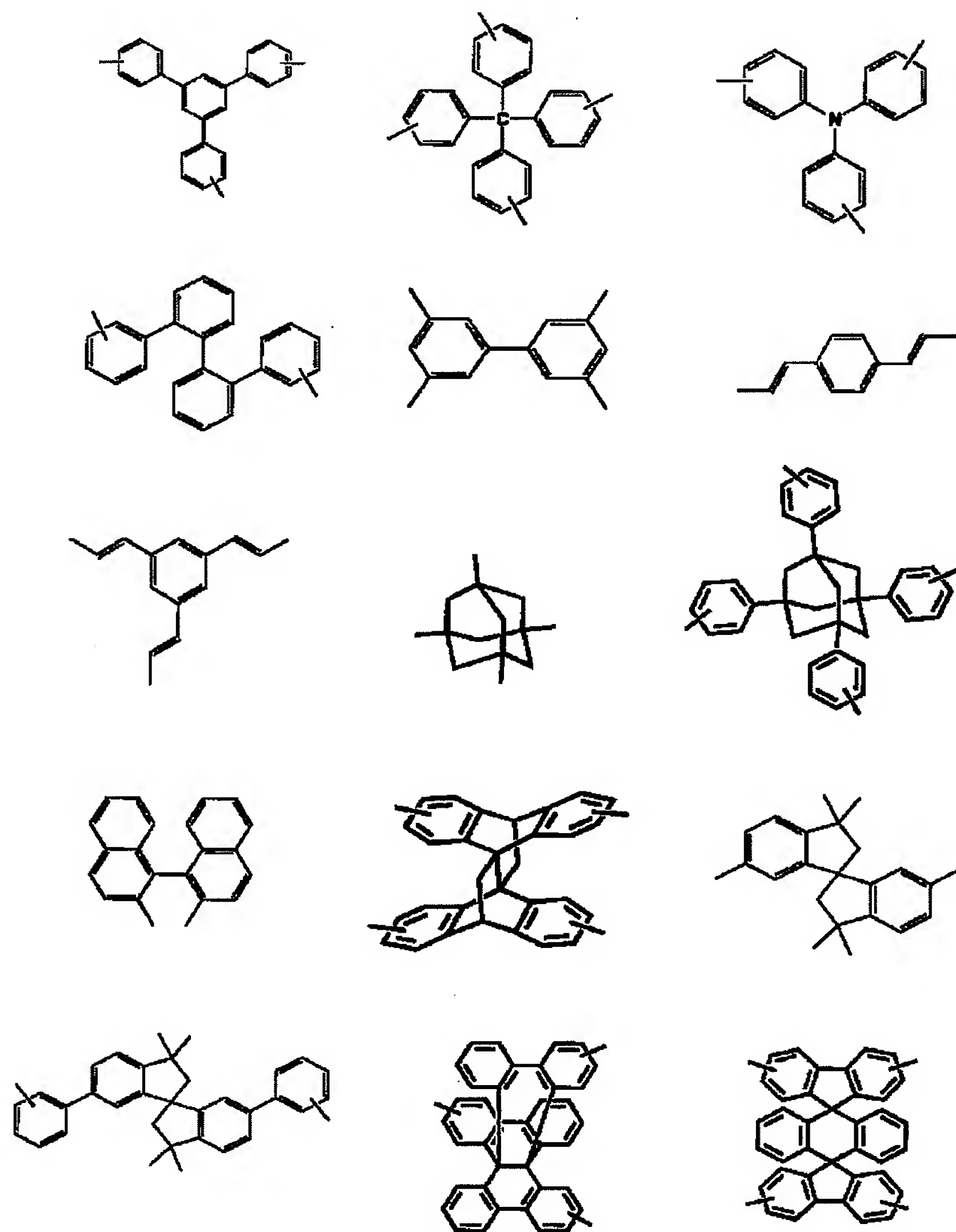
[Formula 14]



[0041]

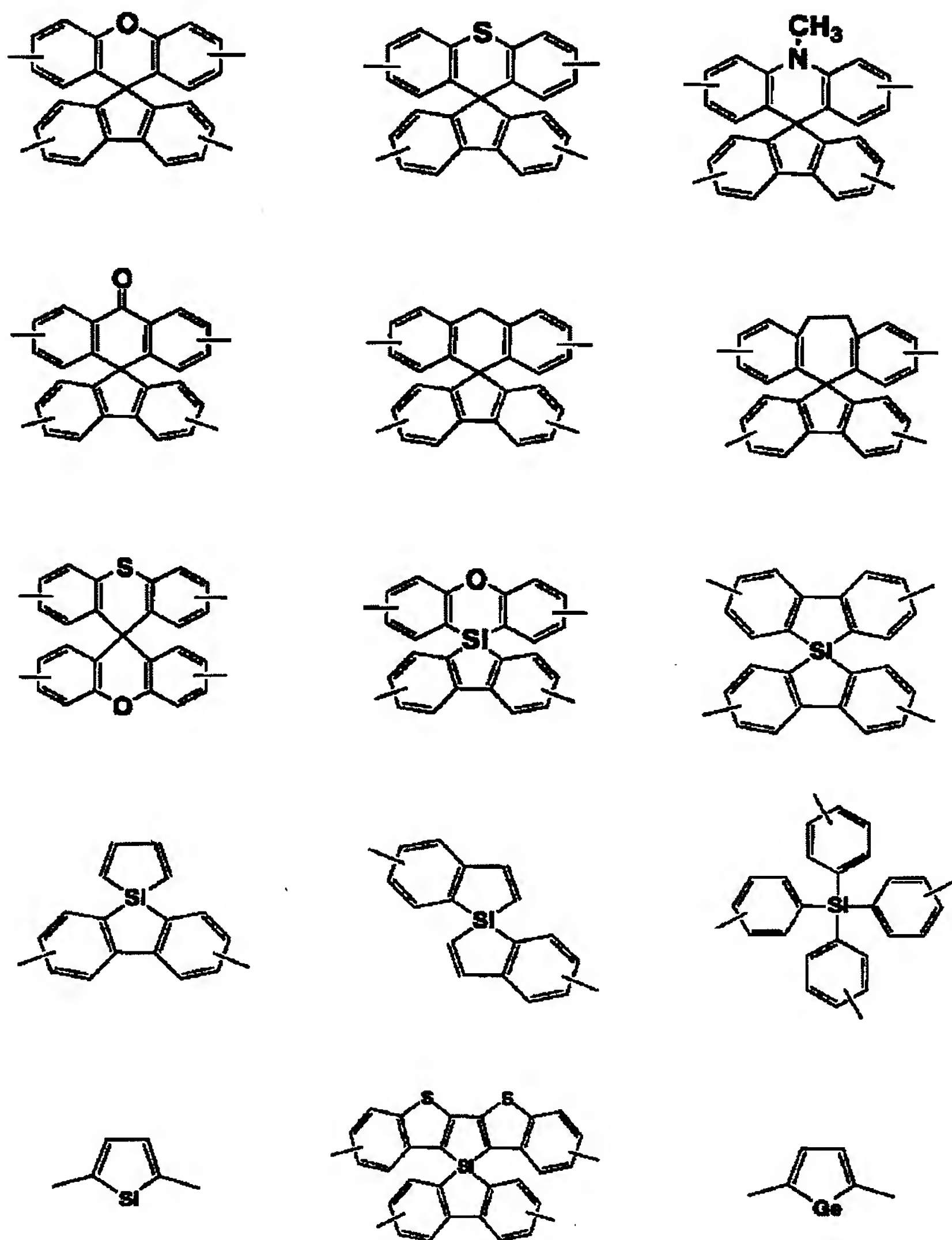
[Formula 15]





[0042]

[Formula 16]



[0043] Although these connecting groups can obtain a commercial thing, or can be compounded in accordance with a conventional method and can form the connected structure, they show the example of concrete composition of some connecting groups below.

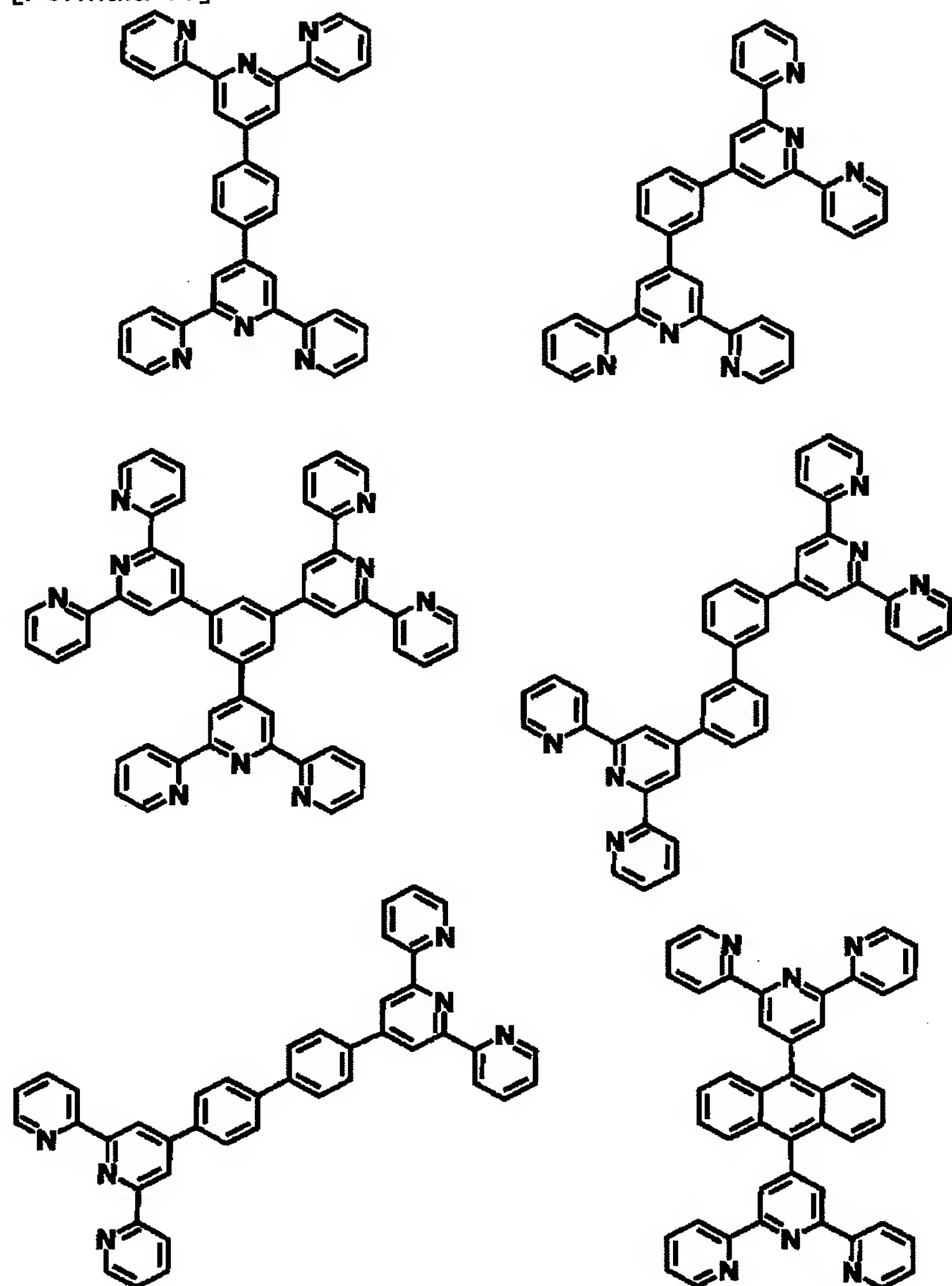
[0044] As for composition of a 9,9-spirobi fluorene skeleton, the 2881st page of J. Am. Chem. Soc. and vol.52 (1930), the example "A. composition of a starting compound (a) 9,9'-spirobifluorene" of JP,7-278537,A, etc. are given. Use 2-bromobiphenyl as a Grignard reagent using metal magnesium in THF, subsequently it is made to react to 9-fluorenone at 50 \*\* from a room temperature, heating drying of the hydroxy \*\* obtained by processing with a conventional method is carried out in the acetic acid which added a small amount of chlorides, and it processes with a conventional method.

[0045] About composition of a tetraphenylmethane skeleton. The 1098th page of Angew.Chem. Int.Ed. Engl. and vol.25(1986)No.12, the 1487th page of Tetrahedron Letters and vol.38 (1997), etc. are mentioned as a reference. Specifically triphenylmethanol or triphenyl methyl chloride among a non-solvent or an acetic acid solvent, Make it react at aniline or an aniline hydrochloride, 100 \*\*, or 220 \*\*, process and isolate with a conventional method and subsequently an obtained intermediate is made to react to an isoamyl night light at -10 \*\* among a mixed solvent of ethanol/sulfuric acid, phosphinic acid is added, a heating rotary flow is carried out, and it processes with a conventional method.

[0046] The following structures are mentioned as an example as a desirable example of a compound which has two or more above-mentioned oligo pyridine skeletons.

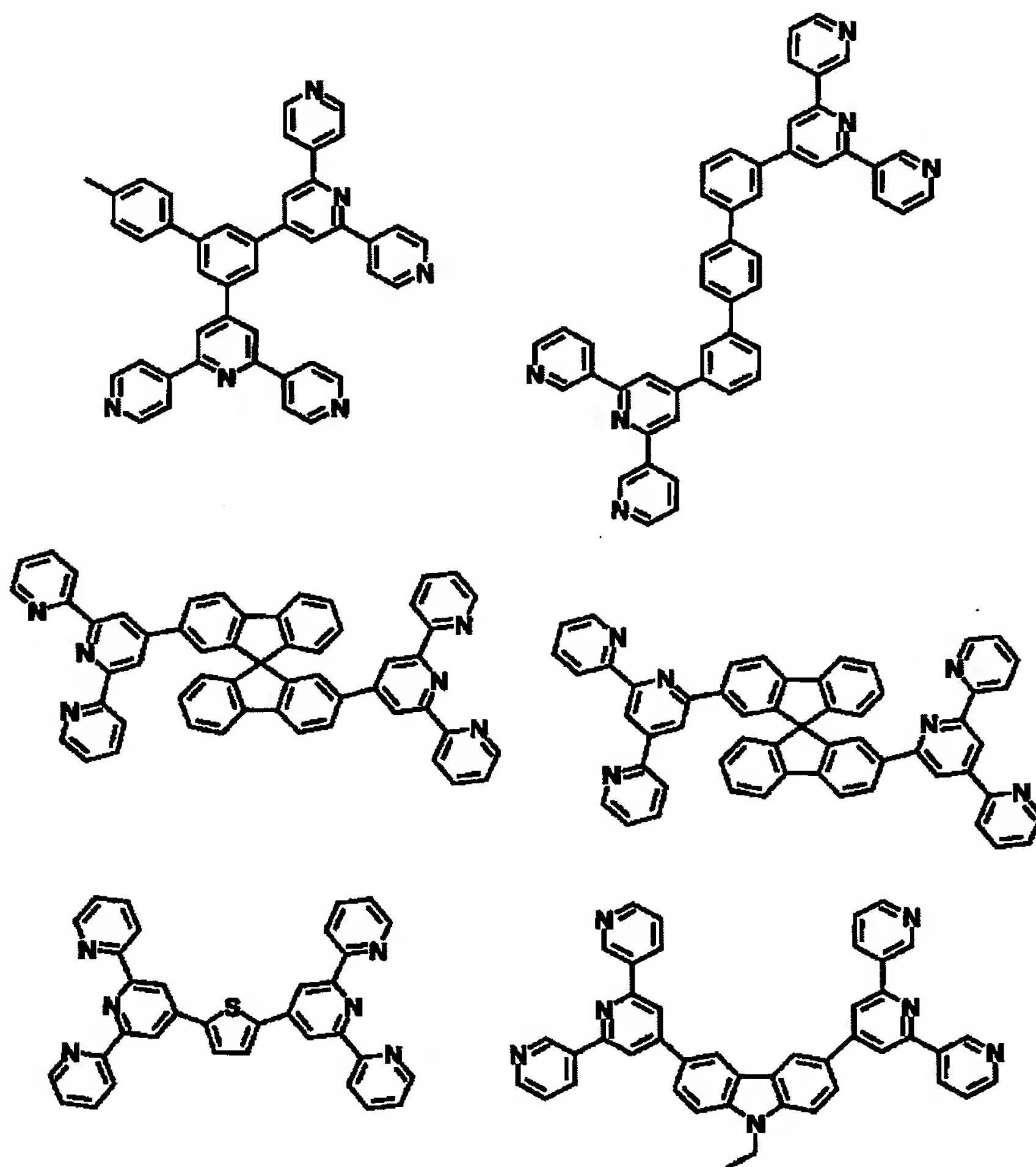
[0047]

[Formula 17]



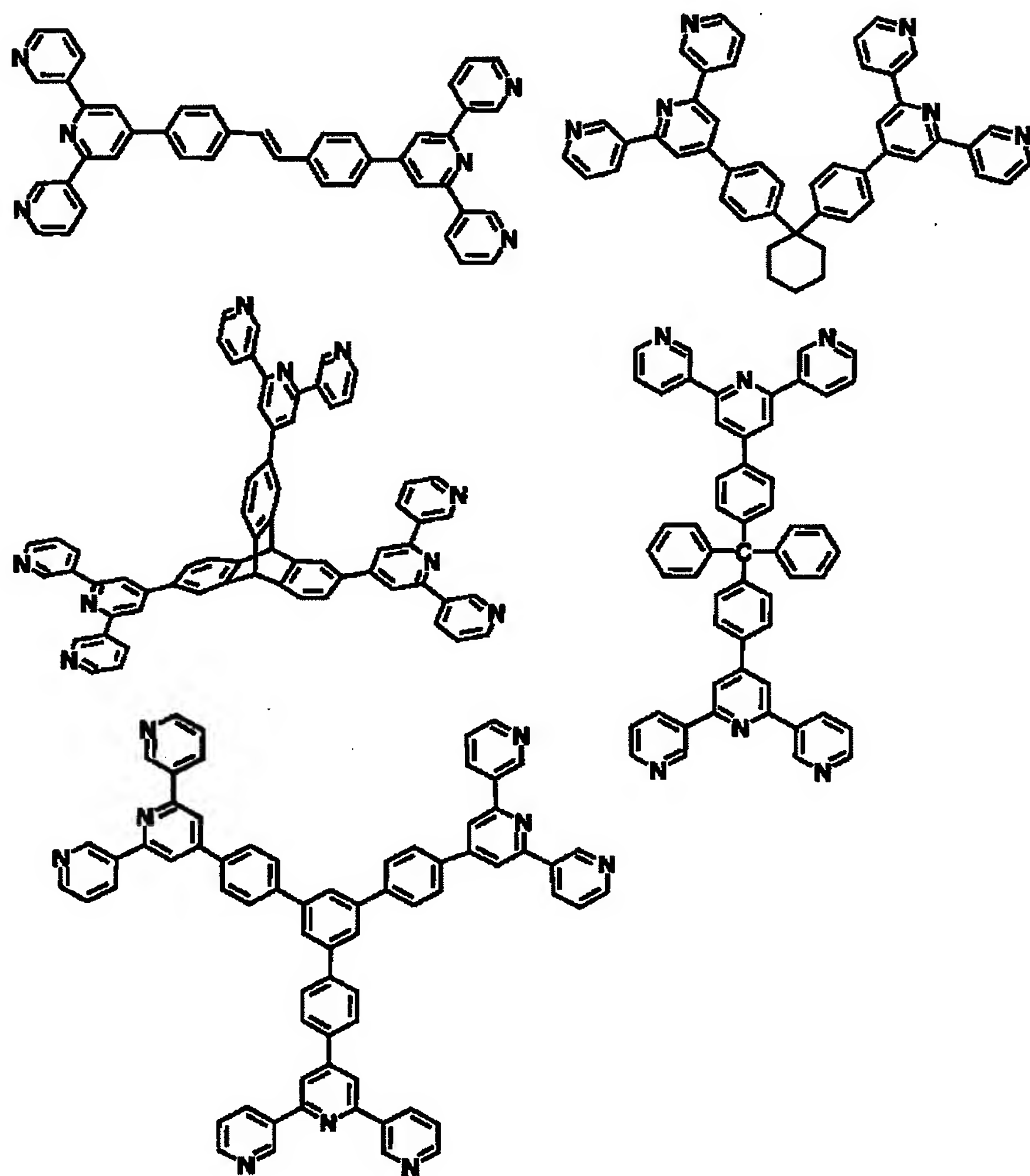
[0048]

[Formula 18]



[0049]

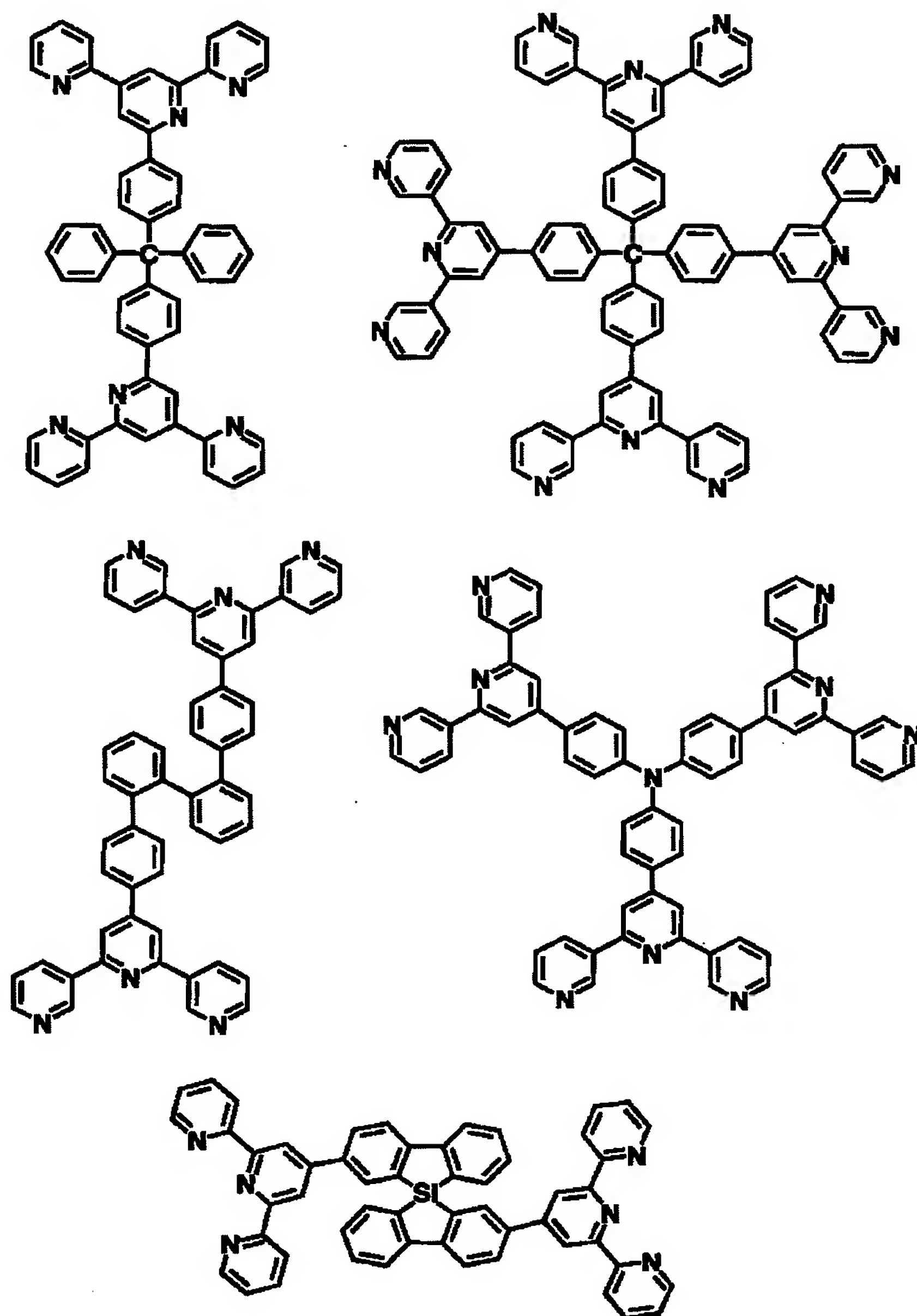
[Formula 19]



[0050]

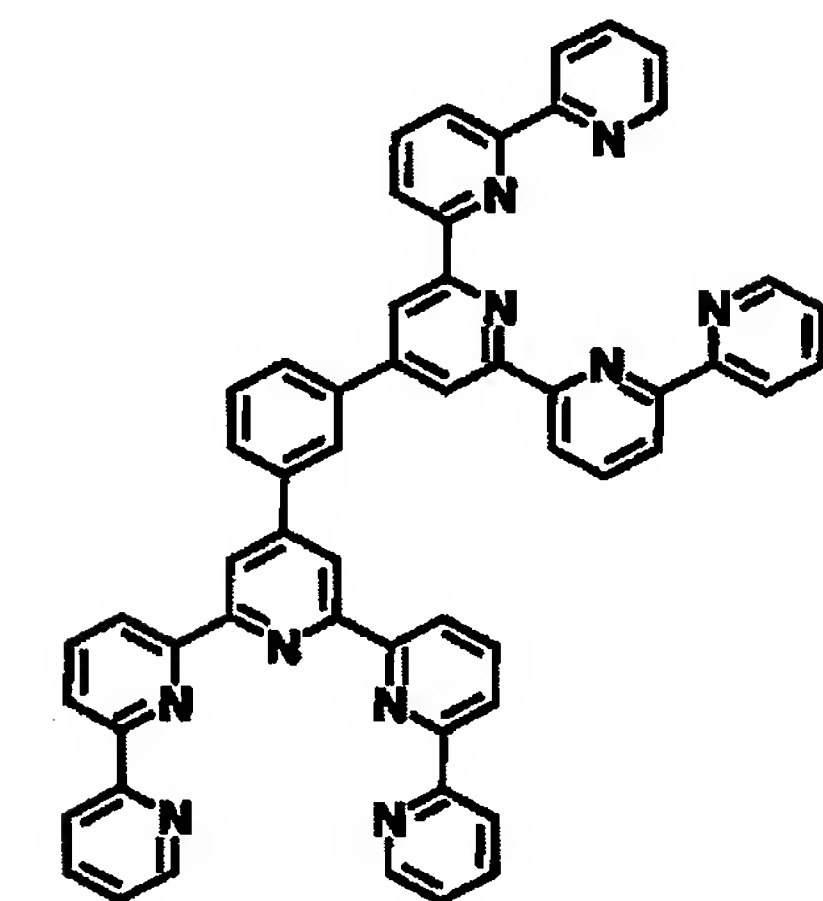
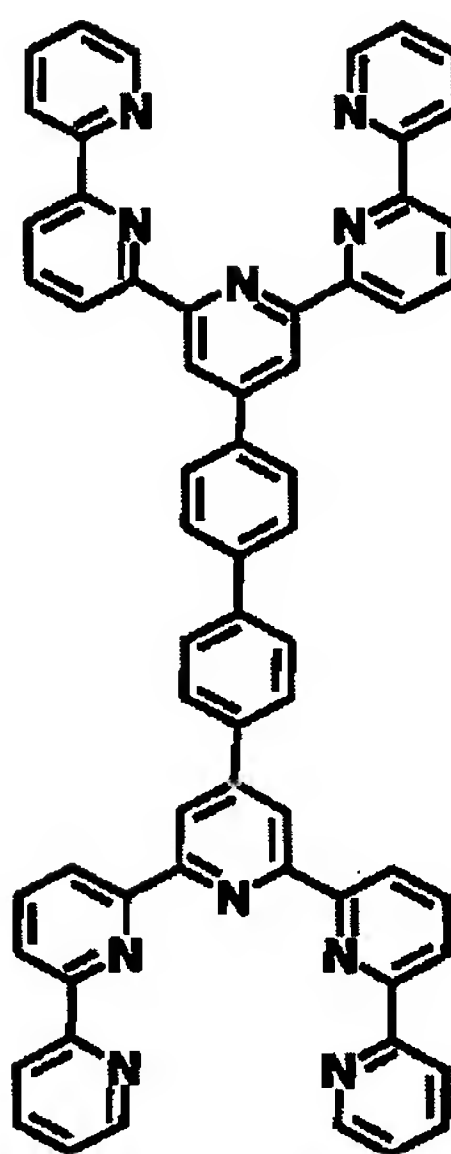
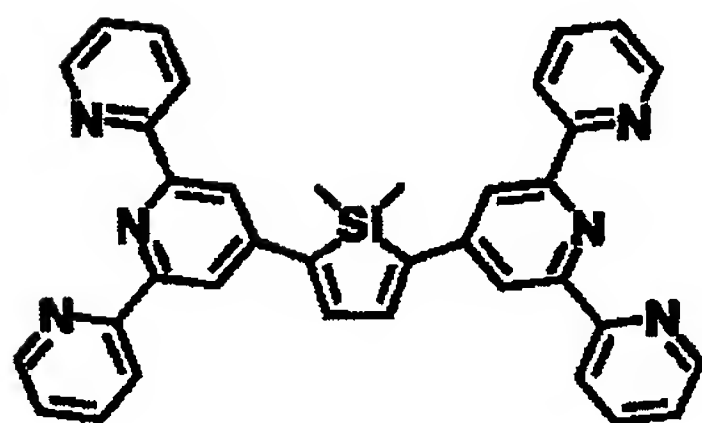
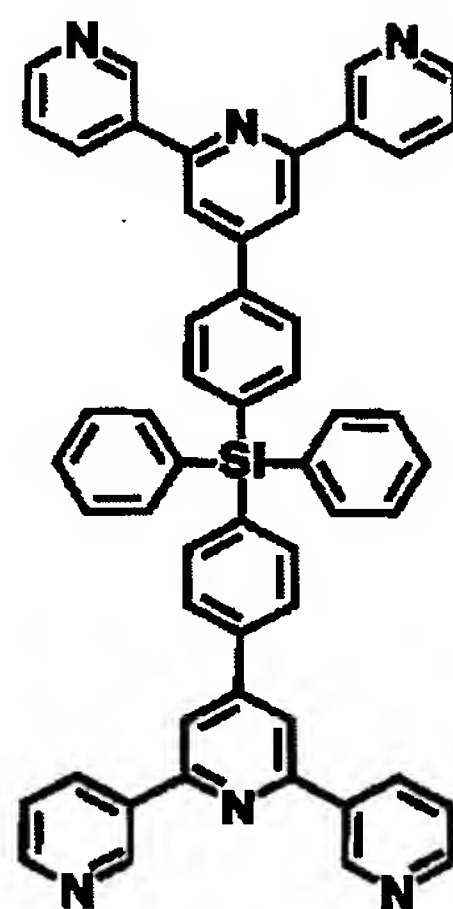
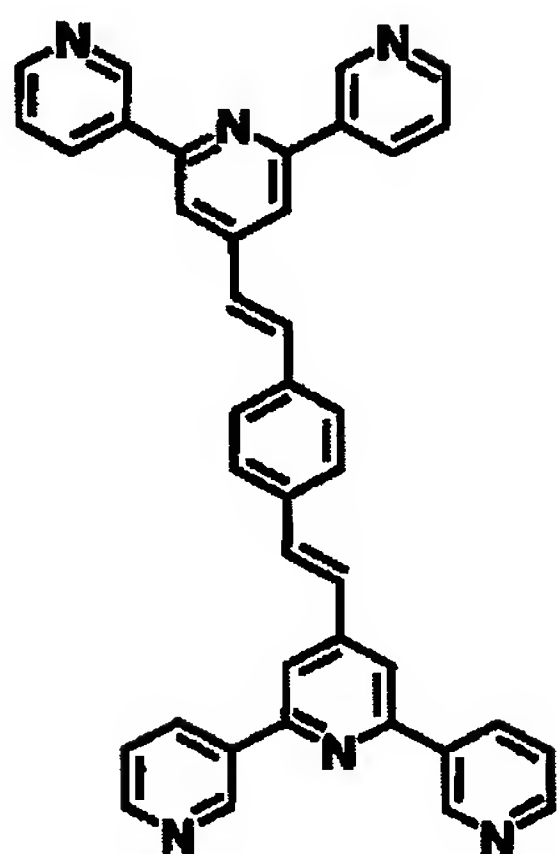
[Formula 20]



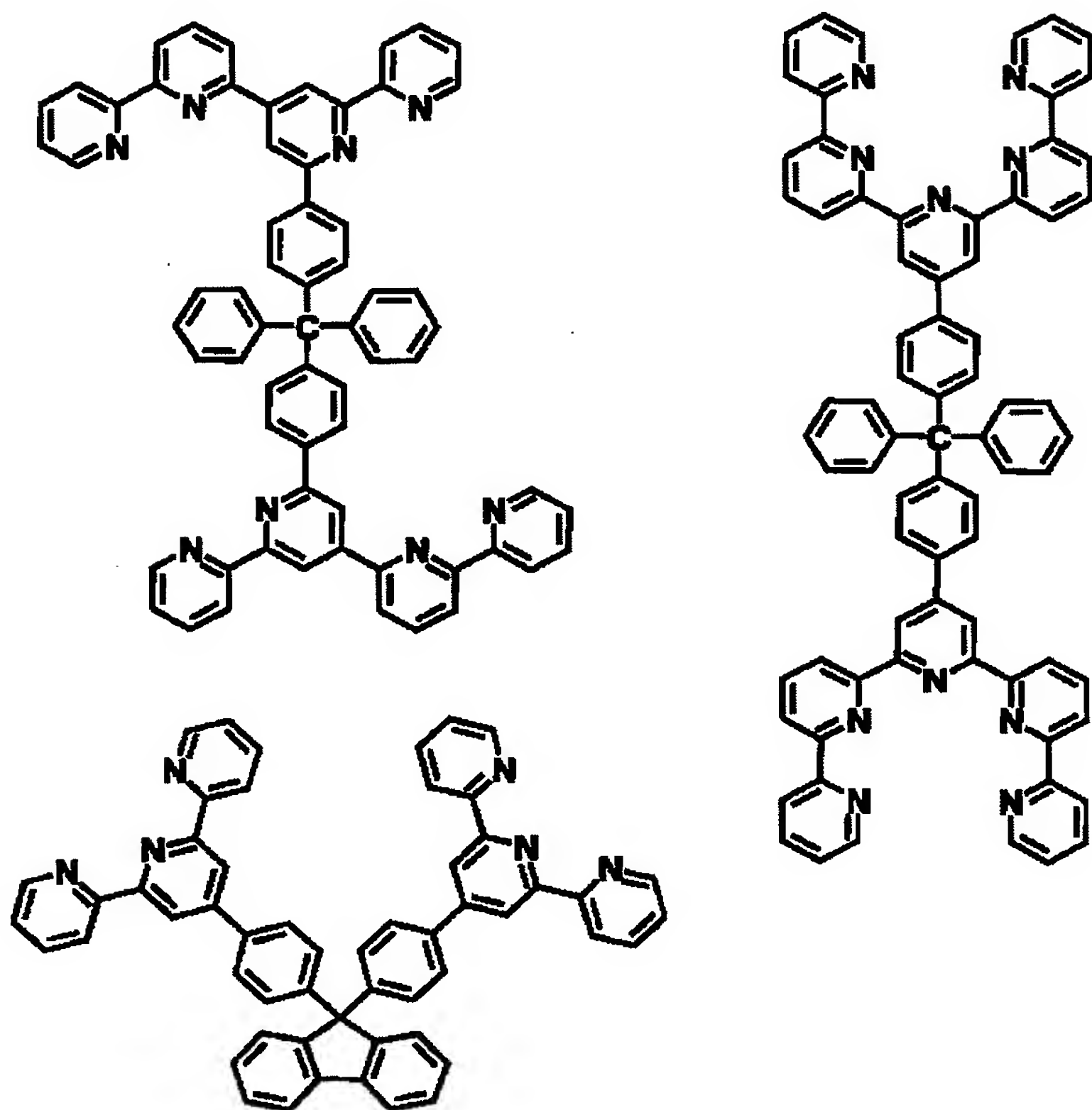


[0051]

[Formula 21]



[0052]  
[Formula 22]



[0053]As an introducing method of the oligo pyridine skeleton to a connecting group, After introducing reactive substituents which form a pyridine ring and are made into an oligo pyridine skeleton after introducing a reactive substituent like a formyl group or an acetyl group, such as a method, an iodo group, and a bromo group, the method of giving an oligo pyridine skeleton is mentioned.

[0054]Acylation of the Friedel crafts general and simple as a method of introducing an acetyl group is raised. As a reference, The 27th page "example A. starting compound (f) 2,2'-diacetyl-9,9'-spirobifluorene" and Helvetica Chimica Acta of JP,7-278537,A, and vol.52 (1969) 1210th page "Experimenteller. Tell 2 and 2'-diacetyl-9,9'-spirobifluorene(IV)" etc. are raised. A connecting group can be made to be able to react to an acetyl chloride and an aluminium chloride at 50 °C in 1,2-dichloroethane, it can process with a conventional method, and an acetyl group can be introduced.

[0055]About introduction of an iodo group, the above-mentioned Tetrahedron Letters, the 1487th page of vol.38 (1997), etc. are raised as a reference. A connecting group can be made to be able to react to iodine and a bis(trifluoroacetoxy)iodobenzene at 50 °C thru/or 60 °C in a carbon tetrachloride, it can process with a conventional method, and an iodo group can be introduced.

[0056]About introduction of a bromo group, the 1098th page of above-mentioned Angew. Chem.Int. Ed. Engl. and 25(1986)No.12, etc. are raised as a reference. A connecting group can be made to be able to react to bromine at a room temperature, it can process with a conventional method, and a bromo group can be introduced.

[0057]An iodo group or a bromo group of a connecting group is made to react to oligo pyridine which RICHIO-izes and subsequently corresponds with metal lithium as introduction of an oligo pyridine skeleton from an iodo group and a bromo group, and there is a method of processing by water and manganese dioxide.

[0058]About introduction of a formyl group, it can obtain by making boron acids, such as formyl phenylboronic acid, react to an iodo group or a bromo group on condition of Suzuki coupling (reference: the 2457th page of Chem.Rev. and vol.95 (1995)). An acetyl group can also be introduced in a similar way.

[0059]Next, a light emitting device using a compound which has an oligo pyridine skeleton of this invention as a charge of light emitting device material is explained in detail.

[0060]In order that the anode may take out light, if transparent, tin oxide, indium oxide, Conductive metallic oxide, such as tin oxide indium (ITO), or gold, Especially things for which ITO glass and Nesa glass are used although not limited, such as especially conductive polymers, such as inorganic conductive substances, such as metal, such as silver and chromium, copper iodide, and copper sulfide, a polythiophene, polypyrrole, and poly aniline, are desirable. Since the resistance of a transparent electrode just supplies sufficient current for luminescence of an element, it is not limited, but from a viewpoint of power consumption of an element, it is desirable that it is low resistance. For example, if it is an ITO board below 300ohms / \*\*, it will function as an element electrode, but since supply of a substrate about 10ohms / \*\* is also attained now, especially a thing for which a low resistance article is used is desirable. Although thickness of ITO can be arbitrarily chosen according to resistance, it is usually used among 100-300 nm in many cases. Since the glass substrate should just have sufficient thickness for soda lime glass, alkali free glass, etc. to be used, and for thickness also maintain a mechanical strength, it is enough if there is 0.5 mm or more. About construction material of glass, since the direction with little elution ion from glass is good, alkali free glass is more preferred, but since soda lime glass which gave barrier coating sealant, such as  $\text{SiO}_2$ , is also marketed, this can be used. As long as the anode functions stably, the substrate does not need to be glass, for example, may form the anode on a plastic plate. An ITO film formation method in particular does not receive restriction an electron beam method, sputtering process, a chemical reaction method, etc.

[0061]Especially if the negative pole is a substance which can pour an electron into this organic layer efficiently, it will not be limited, but generally platinum, gold, silver, copper, iron, tin, zinc, aluminum, indium, chromium, lithium, sodium, potassium, calcium, magnesium, etc. are raised. In order to gather electron injection efficiency and to raise an element characteristic, an alloy containing lithium, sodium, potassium, calcium, magnesium, or these low work function metals is effective. However, although these low work function metals can be mentioned as an example with a preferred way that it is generally unstable in the atmosphere dopes a small amount of lithium and magnesium (it is 1 nm or less by thickness gage display of vacuum deposition) to an organic layer, and it uses many [ for example, ] extremely stable electrodes for it, Use of mineral salt like lithium fluoride is not limited to these in particular from a possible thing, either. For electrode protection, metal, such as platinum, gold, silver, copper, iron, tin, aluminum, and indium, Or it is mentioned as an example with preferred laminating inorganic substances, such as an alloy using these metal and silica, a titania, and silicon nitride, polyvinyl alcohol, VCM/PVC, hydrocarbon system polymers, etc. It will not be restricted especially if a method of producing these electrodes can also take flows, such as resistance heating, an electron beam, sputtering, ion plating, and coating.

[0062]Although photogene contained in a light emitting device of this invention helps what emits light itself, and its luminescence, it corresponds to all, and it points out a compound which is participating in luminescence. Specifically, a hole transporting material, a luminescent material, an electron transport material, etc. correspond. light emitting devices of this invention may be any of a gestalt which was formed of a layer which consists of photogene, boiled further 1 electron-hole transporting bed / luminous layer, 2 electron-hole transporting bed / luminous layer / electron transport layer, 3 luminous layers / electron transport layer, and a combination substance more than four, and was mixed. That is, as element composition, it is also good like 4 to provide further a layer containing a luminescent material independent or a luminescent material, a hole transporting material, and an electron transport material other than a multilayer laminated structure of the above 1-3.

[0063]although being contained as an electron transport material is preferred as for a compound which has an oligo pyridine skeleton of this invention, it is not limited in particular and may be contained in any of the above-mentioned photogene -- much more -- or it may be contained in two or more layers. It may be contained in a part of layer even if contained in the whole layer.

[0064]An electron hole transporting bed laminates a kind of a hole transporting material, or two sorts or more, and is mixed, or is formed with a mixture of a hole transporting material and a



macromolecular binding agent. As a hole transporting material, N,N'-diphenyl-N,N'-JI (3-methylphenyl)-4,4'-diphenyl-1,1'-diamine, Triphenylamines, such as N,N'-dinaphthyl N,N'-diphenyl-4,4'-diphenyl-1,1'-diamine. A screw (N-allylcarbazole) or screws (N-alkyl carbazole). A pyrazoline derivative, a stilbene compound, a hydrazone system compound, an oxadiazole derivative and a phthalocyanine derivative, A heterocyclic compound represented by porphyrin derivative, polycarbonate and a styrene derivative which have said monomer in a side chain in a polymer system, Although a polyvinyl carbazole, polysilane, etc. are preferred, a thin film required for element production is formed, and an electron hole can be poured in from the anode, and it will not be limited especially if it is a compound which can convey an electron hole further.

[0065]A luminous layer may be formed with a luminescent material (a host material, a dopant material), this may be a mixture of a host material and a dopant material, or may be host material independent, or any may be sufficient as it. The number of a host material and dopant materials may be one, respectively, they may be two or more combination, or any may be sufficient as them. A dopant material may be included in the whole host material, may be included selectively, or may be any. It may laminate, or may distribute or dopant materials may be any. If there is too much quantity of a dopant material, in order that a concentration-quenching phenomenon may occur, it is 2 or less % of the weight to use at 10 or less % of the weight to a host material desirable still more preferably. As the doping method, although it can form with vapor codeposition with a host material, after mixing with a host material beforehand, it may vapor-deposit simultaneously.

[0066]As a luminescent material, it can choose from various materials according to the desired luminescent color. Naphthalene which it is not limited in particular in order to obtain bright luminescence, but was known as a photogen for some time, Anthracene, phenanthrene, pyrene, triphenylene, perylene, Aromatic hydrocarbon compounds and derivatives of those, such as TORAKISEN, a fluorene, indene, and a 9,9'-spirobifluorene, A franc, pyrrole, a thiophene, silole, 9-silafluorene, 9,9'-spirobi silafluorene, Benzothiophene, benzofuran, Indore, a dibenzo thiophene, A dibenzofuran, imidazopyridine, a phenanthroline, pyrazine, Aromatic heterocyclic compounds and derivatives of those, such as NAFUCHI lysine, quinoxaline, pyrrolo pyridine, and thoxanthene, Quinolinol metal complexes, such as a tris (8-quinolinolato) aluminium complex, A bipyridine metal complex, a rhodamine metal complex, an azomethine metal complex, a JISUCHIRIRU benzene derivative, A tetraphenyl butadiene derivative, a stilbene derivative, an aldazine derivative, A coumarin derivative, a phthalimide derivative, a NAFUTARU imide derivative, a perinon derivative, a pyrrolo pyrrole derivative, a cyclopentadiene derivative, an acridone derivative, imidazole, a thiazole, thiadiazole, An azole derivative and its metal complexes, such as carbazole, oxazol, oxadiazole, and triazole, A PIROMETEN derivative, a merocyanine derivative, a porphyrin derivative, a PIROMETEN compound and its metal complex, a phosphorescence nature metal complex that used as a central metal iridium represented by tris(2-phenylpyridine) iridium (III) and platinum, etc. can be used conveniently.

[0067]A compound which has an oligo pyridine skeleton of this invention can be similarly used as a luminescent material from having fluorescent luminescence.

[0068]An electron transport layer is a layer which manages an electron being poured in from the negative pole and conveying an electron further, electron injection efficiency is high and it is desirable to convey a poured-in electron efficiently. However, in mainly playing a role which can prevent efficiently flowing into the negative pole side, without an electron hole from the anode recombining when transportation balance of an electron hole and an electron is considered, even if electron transportation capability is not so high, it has an effect of raising luminous efficiency, on a par with material with high electron transportation capability. Therefore, a hole blocking layer from which the electron transport layer in this invention can prevent movement of an electron hole efficiently is also contained as a thing of homonymy.

[0069]An electron transport material in this invention contains a compound which has the oligo pyridine skeleton which connected a pyridine ring expressed with a general formula (1) in or more three single bond.

[0070]In order to obtain stable luminescence over a long time, a material excellent in thermal stability or thin-film-forming nature is desired, and a compound which has two or more oligo



pyridine skeletons also in a compound which has an oligo pyridine skeleton can be raised as a more desirable example.

[0071]In order to obtain bright luminescence using a compound in this invention, it is preferred that electron transportation ability uses a high compound. Then, as a compound which has two or more oligo pyridine skeletons, a compound which includes aromatic heterocycle which is not replaced [ aromatic hydrocarbon which is not replaced / a conjugated bond substitution, or /, substitution, or ] in a bonding unit is more preferred.

[0072]It is not necessary to restrict an electron transport material only to a compound kind which has an oligo pyridine skeleton, and said two or more compounds may be mixed and used, or it may mix one or more kinds of a known electron transport material with said compound, and may be used. Although not limited especially as a known electron transport material, electron injection efficiency is high, It is desirable that a poured-in electron is conveyed efficiently or movement of an electron hole from the anode can be prevented efficiently, A quinolinol derivative metal complex specifically represented by 8-hydroxy kino RINARU minium, A TAPI lysine metal complex, a tropolone metal complex, a flavonol metal complex, A perylene derivative, a peri non derivative, naphthalene, a coumarin derivative, a benzimidazole derivative, A benzoxazole derivative, a benz thiazole derivative, an oxadiazole derivative, A thiadiazole derivative, a triazole derivative, an aldazine derivative, a screw styryl derivative, a pyrazine derivative, a phenanthroline derivative, a quinoxaline derivative, a benzoquinoline derivative, a bipyridine derivative, aromatic phosphorus oxide, etc. are raised.

[0073]The above electron hole transporting bed, a luminous layer, and an electron transport layer are independent, or two or more kinds of materials are laminated, Mix or as a macromolecular binding agent Polyvinyl chloride, polycarbonate, Polystyrene, poly (N-vinylcarbazole), polymethylmethacrylate, Polybutyl methacrylate, polyester, polysulfone, polyphenylene oxide, Polybutadiene, hydrocarbon resin, ketone resin, phenoxy resin, Pori Sall John, Solvent solubility resin, such as polyamide, ethyl cellulose, vinyl acetate, ABS plastics, and polyurethane resin, It is also possible to distribute hardening resin, such as phenol resin, xylene resin, petroleum resin, a urea resin, melamine resin, unsaturated polyester resin, alkyd resin, an epoxy resin, and silicone resin, etc., and to use.

[0074]Although a formation method in particular of a layer which constitutes an element is not limited [ coating method / resistance heating vacuum evaporation, electron beam evaporation, sputtering, a molecule laminated layers method, ], it is usually preferred in respect of the characteristic. [ of resistance heating vacuum evaporation and electron beam evaporation ] Since thickness of a layer is based also on resistance of photogene, it cannot be limited, but it is chosen from for 1-1000 nm.

[0075]Although electrical energy mainly refers to a direct current, it is also possible to use pulse current and alternating current. Although a current value and a pressure value do not have restriction in particular, when power consumption of an element and a life are taken into consideration, the luminosity maximum with the lowest possible energy should be made to be obtained.

[0076]A pixel for a display says things by which it has been arranged in two dimensions, such as the shape of a lattice, and mosaic shape, and a matrix in this invention displays a character and a picture by set of a pixel. Shape of a pixel and size are decided by a use. For example, a pixel with a quadrangle of 300 micrometers or less will usually be used for a picture and a character representation of a personal computer, a monitor, and television for one side, and, in the case of a large sized display like a display panel, a pixel of mm order of one side will be used for them. In the case of a monochrome display, a pixel of the same color should just be arranged, but red, red, and a green and blue pixel are displayed [ a case where it is a colored presentation ]. In this case, there are a delta type and a stripe type typically. And as a drive method of this matrix, either a line sequential drive method or an active matrix is OK. Although there is an advantage that structure is simpler for the line sequential drive, since a direction of an active matrix may be excellent when an operating characteristic is taken into consideration, it is required to also use this properly by a use.

[0077]A field which formed a pattern and was decided to display information beforehand decided

to be a segment type in this invention is made to emit light. For example, an operating state display of time in a digital clock or a thermometer, a temperature display and audio equipment, an induction heating cooker, etc., a panel display of a car, etc. are raised. And said matrix display and a segment display may live together in the same panel.

[0078]A light emitting device of this invention is preferably used also as a back light. A back light is used for the purpose of raising the visibility of a display which does not mainly carry out spontaneous light, and is used for a liquid crystal display, a clock, audio equipment, an automobile panel, the plotting board, a sign, etc. Since a thing of a conventional system consists of a fluorescent lamp or a light guide plate as a back light of a personal computer use in which slimming down has especially been SUBJECT also in a liquid crystal display, if it considers that slimming down is difficult, as for a back light using a light emitting device in this invention, a thin shape and a light weight will become the feature.

[0079]

[Example]This invention is not limited by these examples, although an example and a comparative example are given and this invention is explained hereafter.

[0080]5 g of isophthalaldehyde, 9 g of 2-acetylpyridine, and the potassium hydrate 5.3g were put in 30 ml of synthesizing method ethanol of example 1, 1,3-bis(2,6-dipyridyl pyridin-4-yl)benzene, and the mixed solution of 32 ml of water, and it was made to react at a room temperature for 2 hours. Vacuum drying was washed and carried out by water and ethanol after filtration. The obtained white powder 1g was dissolved in 30 ml of ethanol, 1.9 g of 2-PIRIDA sill pyridiniumiodide and the ammonium acetate 12g were added, and the heating rotary flow was carried out for 4 hours. After it filtered after cooling to a room temperature, and methanol washed, recrystallization was performed twice by dimethylformamide and the 1,3-bis(2,6-dipyridyl pyridin-4-yl)benzene 0.5g was obtained. The  $^1\text{H-NMR}$  analysis result of the obtained powder was as follows.

$^1\text{H-NMR}(\text{CDCl}_3 \text{ (d=ppm)})$ : 7.31–7.39 (m, 4H), 7.65 (q, 1H), 7.84–8.00 (m, 6H), 8.32 (s, 1H), and 8.66–8.87 (m, 12H) — subsequently the light emitting device using 1,3-bis(2,6-dipyridyl pyridin-4-yl)benzene was produced as follows. Cutting and etching were performed for the glass substrate (the Asahi Glass Co., Ltd. make, 15ohm/\*\*, electron-beam-evaporation article) on which 150 nm of ITO transparent conducting films were made to deposit to 30x40 mm. After cleaning the obtained substrate ultrasonically for 15 minutes respectively by acetone and the "semi-coculine 56" (made by Furuuchi Chemical Corp.), ultrapure water washed. Then, heat methanol was made to immerse for 15 minutes after cleaning ultrasonically for 15 minutes with isopropyl alcohol, and it was made to dry. Just before producing an element for this substrate, it is 1-hour UV. — It ozonized and installed in the vacuum evaporator, and it exhausted until the degree of vacuum in a device became below  $5 \times 10^{-5} \text{ Pa}$ . With the resistance heating method, first, the copper phthalocyanine was vapor-deposited as a hole-injection material, and 100 nm of 4,4'-bis(N-(1-naphthyl)-N-phenylamino)biphenyls were vapor-deposited as 20 nm and a hole transporting material. Next, as a luminescent material, the trisquinolinol aluminium complex (Alq3) was laminated in thickness of 50 nm. Next, as an electron transport material, the above-mentioned 1,3-bis(2,6-dipyridyl pyridin-4-yl)benzene was laminated in thickness of 100 nm. Next, after doping lithium to a 0.5-nm organic layer, 200 nm of aluminum was vapor-deposited, it was considered as the negative pole, and the element of the 5x5 mm square was produced. The thickness said here is a crystal oscillation type thickness value monitor value. With the luminous wavelength of 530 nm and a luminosity of 19000 cds/square meter high-intensity green emission was obtained from this light emitting device. The endurance of this light emitting device was dramatically excellent, and held not less than 70% of luminosity in 1000 hours.

[0081]Good luminescence was checked when carrying out the 1-mA pulse drive (the Duty ratios 1/60, current value of 60 mA at the time of a pulse) of this light emitting device within the vacuum cell.

[0082]The element was produced like Example 1 except having used 1,4-bis(2,6-dipyridyl pyridin-4-yl)benzene as example 2 electron transport material. With the luminous wavelength of 532 nm and a luminosity of 16000 cds/square meter high-intensity green emission was obtained

from this light emitting device. The endurance of this light emitting device was dramatically excellent, and held not less than 70% of luminosity in 800 hours.

[0083]The 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BTCNP) was used as comparative example 1 electron transport material, and also the light emitting device was produced completely like Example 1. The endurance of this light emitting device was remarkably short, and luminosity reduced it by half in 100 hours.

[0084]As example 3 luminescent material, as a host material, a trisquinolinol aluminium complex (Alq3), As a guest material, 4-(dicyanomethylene)-2-*t*-butyl-6-(1,1,7, and 7-tetramethyl-9-enyl)-4H-Piran (DCJTB) was used so that dope concentration might be 2%, and also the light emitting device was produced completely like Example 1. With the luminous wavelength of 630 nm and a luminosity of 9000 cds/square meter high-intensity red lamp color luminescence was obtained from this light emitting device. The endurance of this light emitting device was dramatically excellent, and held not less than 70% of luminosity in 1000 hours.

[0085]As example 4 host material, 4,4'-bis(diphenylvinyl)biphenyl (DPVBi) was used, and also the light emitting device was produced completely like Example 1. With the luminous wavelength of 460 nm and a luminosity of 8000 cds/square meter high-intensity blue light was obtained from this light emitting device.

[0086]The trisquinolinol aluminium complex (Alq3) was used as comparative example 2 electron transport material, and also the light emitting device was produced completely like Example 4. In addition to luminescence from a luminescent material, from this light emitting device, the green emission from an electron transport material was observed, and color purity was remarkably bad.

[0087]As example 5 host material, 1,4-diketo 2,5-bis(3,5-dimethylbenzyl)-3,6-bis(4-methylphenyl)pyrrolo [3,4-*c*] pyrrole, As a guest material, 4,4-difluoro-1,3,5,7-tetra (4-*n*-buthylphenyl)-8-phenyl-4-\*\*\*\*-3a,4a-diaza-*s*-indacene was used so that dope concentration might be 1%, and also the light emitting device was produced completely like Example 1. With the luminous wavelength of 616 nm and a luminosity of 15000 cds/square meter high-intensity red light was obtained from this light emitting device.

[0088]The glass substrate (the Asahi Glass Co., Ltd. make, 15ohm/\*\*, electron-beam-evaporation article) on which 150 nm of example 6ITO transparent conducting films were made to deposit was cut to 30x40 mm, and pattern processing was carried out to 300micrometer pitch (remaining 270 micrometers in width) x32 stripe shape with photolithography method. Long side direction one side of the ITO stripe is extended to a 1.27-mm pitch (opening 800 micrometers in width), in order to make an electrical link with the exterior easy. After cleaning the obtained substrate ultrasonically for 15 minutes respectively by acetone and the "semi-coculine 56", ultrapure water washed. Then, heat methanol was made to immerse for 15 minutes after cleaning ultrasonically for 15 minutes with isopropyl alcohol, and it was made to dry. Just before producing an element for this substrate, it is 1-hour UV. - It ozonized and installed in the vacuum evaporator, and it exhausted until the degree of vacuum in a device became below  $5 \times 10^{-4}$  Pa. With the resistance heating method, 150 nm of 4,4'-bis(N-(*m*-tolyl)-N-phenylamino)biphenyls were first vapor-deposited as a hole transporting material, and the trisquinolinol aluminium complex was vapor-deposited in thickness of 50 nm as a luminescent material. Next, as an electron transport material, 1,3-bis(2,6-dipyridyl pyridin-4-yl)benzene was laminated in thickness of 100 nm. The thickness said here is a crystal oscillation type thickness value monitor value. Next, they are 16 250-micrometer openings (remaining 50 micrometers in width.) by wet etching to a 50-micrometer-thick covar board. Mask exchange of the mask fairly provided in a 300-micrometer pitch was carried out so that it might intersect perpendicularly with an ITO stripe in a vacuum, and it fixed with the magnet from the rear face so that a mask and an ITO board might stick. And after doping lithium to a 0.5-nm organic layer, 200 nm of aluminum was vapor-deposited and the 32x16 dot-matrix element was produced. When carrying out matrix driving of this element, the character representation was able to be carried out without the cross talk.

[0089]The trisquinolinol aluminium complex (Alq3) was used as comparative example 3 electron



transport material, and also the light emitting device was produced completely like Example 5. In addition to the red light from a luminescent material, from this light emitting device, the green emission from an electron transport material was observed, and color purity was remarkably bad.

[0090]

[Effect of the Invention] This invention is excellent in thermal stability, and its utilization efficiency of electrical energy is high, and it can provide the light emitting device excellent in color purity.

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[Translation done.]